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HEALTH PHYSICS

The Official Journal of the Health Physics Society

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An abstract of 200 words or less must accompany the manuscript. It should give a precise account of the major results of the research as well as some indication of methods or applications when these are of interest. Avoid excessively general remarks.

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LIST OF CONTENTS

NUMBER 1

Recommendations of the International Commission on Radiological Protection 1

J. D. McLENDON: Accidental radiation excursion at the Oak Ridge Y-12 Plant—II. Health physics aspects of the accident 21

W. H. LANGHAM and E. C. ANDERSON: Cs^{137} biospheric contamination from nuclear weapons tests . . . 30

E. J. STORY and A. W. WALTNER: Measurement of radioactivity in air filters 49

BERGENE KAWIN: Retention and localization of phosphorus-32 in rat ovary 53

E. B. WAGNER and G. S. HURST: Gamma response and energy losses in the absolute fast neutron dosimeter . 57

A. R. SCHULERT, E. J. HODGES, E. S. LENHOFF and J. L. KULP: Strontium-90 distribution in the human skeleton 62

H. W. PATTERSON, W. N. HESS, B. J. MOYER and R. W. WALLACE: The flux and spectrum of cosmic-ray produced neutrons as a function of altitude 69

R. H. RITCHIE: Calculations of energy loss under the bias in fast neutron dosimetry 73

Notes:

D. A. MORKEN: Integral-phosphor counting of α-emitters 77

R. ZENDLE and E. E. GOODALE: Some unusual X-radiation dosimetry problems associated with radar installations 78

News 81

Health Physics Society Notice 86

Abstracts and titles of papers presented at the Fourth Annual Meeting of the Health Physics Society . . 87

Abstracts of papers presented at the Risø Symposium on Technical Methods in Health Physics . . . 100

A list of papers for publication in future issues 119

NUMBER 2

G. S. HURST, R. H. RITCHIE and L. C. EMERSON: Accidental radiation excursion at the Oak Ridge Y-12 Plant—III. Determination of radiation doses 121

G. A. ANDREWS, B. W. SITTERSON, A. L. KRETCHMAR and M. BRUCER: Accidental radiation excursion at the Oak Ridge Y-12 Plant—IV. Preliminary report on clinical and laboratory effects in the irradiated employees. 134

J. B. BEST: Maximum likelihood analysis of ionizing radiation induced mortality in whole body fractionated dose experiments 139

J. B. BEST: Theoretical relationship between percent kill and shortened life span measures of radiation injury 157

J. S. HANDLOSER: Stray radiation dosimetry around pulsed accelerators 165

W. H. LANGHAM: Physiology and toxicology of plutonium-239 and its industrial medical control 172

A. L. BONI: Rapid determination of mixed beta–gamma radionuclides in urine 186

R. GARRETT and J. S. LAUGHLIN: A diagnostic X-ray exposure dose chamber 189

H. L. BUTLER and R. W. VAN WYCK: Integrity of vinyl plastic suits in tritium atmospheres 195

R. GOLDEN and E. TOCHILIN: Characteristic curves from different ionizing radiations and their significance in photographic dosimetry 199

E. C. WATSON: A film technique for measuring the exposure dose from plutonium 207

Note:

J. F. SOMMERS and I. J. WELLS: A fission gas monitor for gas cooled or off-gas experimental systems . . 213

Letters to the Editor:

N. I. SAX, R. J. SHERER and R. T. DREW: Emergency monitoring methods for the determination of the effective bone-seeking fission products in milk 216

P. R. GUINN: Suggested insignia for the Health Physics Society 217

D. C. LAWRENCE: Fabrication of thick homogeneous β sources 217

News 219

A list of papers to be published in future issues 223

NUMBER 3

PATRICIA W. DURBIN: Metabolic characteristics within a chemical family 225

H. MUTH, B. RAJEWSKY, H.-J. HANTKE and K. AURAND: The normal radium content and the Ra^{226}/Ca ratio of various foods, drinking water and different organs and tissues of the human body 239

R. L. BLANCHARD, B. KAHN and R. D. BIRKHOFF: The preparation of thin, uniform, radioactive sources by surface adsorption and electrodeposition 246

R. H. GOEKE and C. L. WEAVER: Health physics field operations in Atomic Energy Commission weapons-testing program 256

C. W. SILL and J. K. FLYGARE, JR.; Iodine monitoring at the National Reactor Testing Station 261

R. E. COFIELD: *In vivo* gamma counting as a measurement of uranium in the human lung 269

A. L. BONI: Urinalysis method for enriched uranium 288

GEORGE W. ROYSTER, JR.: Electrodeposition of uranium from urine 291

S. MARSHALL SANDERS, JR.: Power functions relating excretion to body burden 295

Notes:

RICHARD C. MCCALL: Low-background shields 304

J. NOSEK and V. CHMELAŘ: On the present possibilities of washing radioactive substances off the skin of living animals 306

F. W. SANDERS, J. A. AUXIER and J. S. CHEKA: A simple method of minimizing the energy dependence of pocket ionization chambers 308

Letter to the Editor:

J. C. HART: Suggested insignia for the Health Physics Society 310

News 311

Erratum 316

Addendum to ICRP Publication 1 317

NUMBER 4

A. E. BOYD and E. E. MORRIS: Spatial distribution of energy dissipated by fallout β-rays 321

H. FOREMAN, W. MOSS and W. LANGHAM: Plutonium accumulation from long-term occupational exposure . 326

R. LIE, R. G. THOMAS and J. K. SCOTT: The distribution and excretion of thallium-204 in the rat, with suggested MPC's and a bio-assay procedure 334

BO LINDELL: An approach to the question of computing doses and effects from fall-out 341

P. E. MORROW: Some physical and physiological factors controlling the fate of inhaled substances—I. Deposition 366

L. J. CASARETT: Some physical and physiological factors controlling the fate of inhaled substances—II. Retention 379

C. T. SCARBORO and L. B. SILVERMAN: A simplified β-ray extrapolation chamber 387

T. TAMURA and D. G. JACOBS: Structural implications in cesium sorption 391

J. B. MCCASLIN: A high-energy neutron-flux detector 399

Note:

M. L. RANDOLPH: The location of lost γ-ray or neutron sources 408

News 410

A list of papers to be published in future issues 418

Erratum 418

AUTHOR INDEX

ANDERSON, E. C. p. 30
ANDREWS, G. A. p. 134
AURAND, K. p. 239
AUXIER, J. A. p. 308
BEST, J. B. pp. 139, 157
BIRKHOFF, R. D. p. 246
BLANCHARD, R. L. p. 246
BONI, A. L. pp. 186, 288
BOYD, A. E. p. 321
BRUCER, M. p. 134
BUTLER, H. L. p. 195
CASARETT, L. J. p. 379
CHEKA, J. S. p. 308
CHMELAR, V. p. 306
COFIELD, R. E. p. 269
DREW, R. T. p. 216
DURBIN, P. W. p. 225
EMERSON, L. C. p. 121
FLYGARE, J. K., JR. p. 261
FOREMAN H. p. 326
GARRETT, R. p. 189
GOEKE, R. H. p. 256
GOLDEN, R. p. 199
GOODALE, E. E. pp. 78, 316
GUINN, P. R. p. 217
HANDLOSER, J. S. p. 165
HANTKE, H.-J. p. 239
HART, J. C. p. 310
HESS, W. N. p. 69
HODGES, E. J. p. 62
HURST, G. S. pp. 57, 121
JACOBS, D. G. p. 391
KAHN, B. p. 246
KAWIN, B. p. 53
KRETCHMAR, A. L. p. 134
KULP, J. L. p. 62
LANGHAM, W. H. pp. 30, 172, 326
LAUGHLIN, J. S. p. 189
LAWRENCE, D. C. p. 217
LENHOFF, E. S. p. 62
LIE, R. p. 334
LINDELL, B. p. 341
MCCALL, R. C. p. 304
MCCASLIN, J. B. p. 399
MCLENDON, J. D. p. 21
MORKEN, D. A. p. 77
MORRIS, E. E. p. 321
MORROW, P. E. p. 366
MOSS, W. p. 326
MOYER, B. J. p. 69
MUTH, H. p. 239
NOSEK, J. p. 306
PATTERSON, H. W. p. 69
RAJEWSKY, B. p. 239
RANDOLPH, M. L. p. 408
RITCHIE, R. H. pp. 73, 121
ROYSTER, G. W., JR. p. 291
SANDERS, F. W. p. 308
SANDERS, S. M., JR. p. 295
SAX, M. I. p. 216
SCARBORO, C. T. p. 387
SCHULERT, A. T. p. 62
SCOTT, J. K. p. 334
SHERER, R. J. p. 216
SILL, C. W. p. 261
SILVERMAN, L. B. p. 387
SITTERSON, B. W. p. 134
SOMMERS, J. F. p. 213
STORY, E. J. p. 49
TAMURA, T. p. 199
THOMAS, R. G. p. 334
TOCHILIN, E. p. 199
WAGNER, E. B. p. 57
WALLACE, R. W. p. 69
WALTNER, A. W. p. 49
WATSON, E. C. p. 207
WEAVER, C. L. p. 256
WELLS, I. J. p. 213
WYCK, R. W. VAN p. 195
ZENDLE, R. pp. 78, 316

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RECOMMENDATIONS OF THE INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION*

PREFACE

THE International Commission on Radiological Protection (ICRP) has been functioning since 1928 when it was established, under the name of International X-ray and Radium Protection Commission, by the Second International Congress of Radiology held in Stockholm, Sweden. It assumed the present name and organizational form in 1950 in order to cover more effectively the rapidly expanding field of radiation protection.

The recommendations published in the present volume represent concepts and practices evolved from discussions at formal and informal meetings of the Commission and its Committees, held in recent years. Prior to World War II the Commission published recommendations at intervals of about three years. The first meeting in the post-war period was held in London in 1950. An informal meeting was held in Stockholm in 1952 in connection with the conference organized by the International Joint Committee on Radiobiology primarily to discuss the genetic effects of radiation. The next meeting was held in Copenhagen in 1953, at which time four of the Committees held formal meetings for the first time. The results of the deliberations were published in 1955 as Supplement No. 6 of the British Journal of Radiology. The Commission and its Committees met again in the spring of 1956 in Geneva, at which time most of the recommendations in the present volume were adopted in principle. At this meeting the Commission became formally affiliated with the World Health Organization (WHO) as a "non-governmental participating organization".

The Commission (ICRP) and the International Commission on Radiological Units (ICRU) held a special joint meeting in New York in the fall of 1956 to consider an invitation from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) for cooperation in the phase of its work involving exposure from medical procedures. The two Commissions accepted the assigned task and submitted a report to UNSCEAR, published in the October 1957 issue of *Physics in Medicine and Biology*. Funds for expenses incurred in the preparation of the report were provided by the UNSCEAR. At the time of this joint meeting the Commission held an informal meeting to discuss further the recommendations to be made following the deliberations in Geneva earlier in the year. Additional discussions were carried out at another special meeting of the Commission held in New York in the spring of 1958. At this time an ad hoc Publication Committee was appointed to expedite the preparation of the manuscripts. This Committee held a two-week meeting in New York in May and prepared a first draft of the recommendations, which was sent out to all members of the Commission. The Committee met again during the second half of July and revised the first draft in accordance with suggestions made by members of the Commission. At this time, the Committee reviewed also the available drafts of the reports of the Commission's Committees.

In the preparation of the Commission's recommendations, the Publication Committee found it necessary to fill certain gaps involving items that had not been formally discussed and approved by the Commission. For this reason advantage was taken of the presence of seven members of the Commission in Geneva in

* Reprinted from *The Report of the Main Commission of the International Commission on Radiological Protection*. Pergamon Press, London, New York (1959).

September 1958 to review the second draft and make appropriate changes. The amendments were sent to the members who could not attend this meeting. The final draft embodies further comments made by members of the Commission.

Many individuals, who are not members of the Commission, were consulted in the preparation of the recommendations and their co-operation is gratefully acknowledged. In particular the recommendations on genetic dose were discussed by the Chairman of the Publication Committee with several prominent geneticists at the Xth International Congress of Genetics in Montreal, August 1958. It should be noted, also, that Committee I at its 1956 meeting in Geneva, prepared the first draft of the new recommendations adopted by the Commission at that time, which provided the basis for the recommendations published in the present volume.

In recent years the Commission has received financial assistance from the International Society of Radiology, the National Association of Swedish Insurance Companies and private Swedish sources for incidental secretarial expenses. Practically all the work of the Commission has been done on a voluntary basis by its members and members of its Committees. The Commission is heavily indebted to them for their efforts and to their parent organizations for technical and secretarial help. The World Health Organization contributed funds for travel expenses of some members of the Publication Committee, for secretarial help and for incidental expenses in the preparation and circulation of the manuscripts. The Commission takes this opportunity to express its deep appreciation of these contributions, without which the preparation of the present recommendations would have been greatly hampered.

The chairman of the Publication Committee and the Temporary Secretary wish to thank the members of the Commission for their cooperative and prompt replies to the numerous questionnaires and ballots that had to be circulated in the course of preparing the final manuscript. Thanks are due also to the members of the Publication Committee who prepared the first two drafts.

The Commission is happy to announce that the Pergamon Press has generously assumed financial responsibility for the publication and distribution (at modest prices and without copyright restrictions) of the present volume and others in preparation embodying the reports of the Commission's Committees.

ROLF M. SIEVERT

Chairman of ICRP

GIOACCHINO FAILLA

Vice-Chairman ICRP
Chairman of Publication Committee

ORGANIZATION

Rules Governing the Selection and Work of the International Commission on Radiological Protection

(1) The International Commission on Radiological Protection (ICRP) functions under the auspices of the International Congress of Radiology. The following rules, amended in 1953 by the International Executive Committee (IEC) of the Congress, govern the selection and work of the ICRP.

I. (a) The International Commission on Radiological Protection (ICRP) shall be composed of a Chairman and not more than 12 members. The selection of members shall be made by the International Executive Committee (IEC) from a list of nominations submitted by the national delegations and by the International Commission on Radiological Protection itself. Members of the ICRP shall be chosen on the basis of their recognized activity in the fields of radiology, radiation protection, physics, biology, genetics, biochemistry, and biophysics, without regard to nationality.

(b) The members of the ICRP shall be selected during one International Congress to serve through the succeeding Congress. Not less than two, but not more than four, members of the ICRP shall be changed at each Congress. In the intervening period a vacancy caused by conditions beyond the control of the IEC shall be filled on the recommendation of the ICRP.

(c) In the event of a member of the ICRP being unable to attend the ICRP meetings, a substitute may be selected by the ICRP as a temporary replacement. Such a substitute member shall not have voting privileges at the meetings unless specifically authorized by the IEC.

(d) The ICRP shall be permitted to invite individuals to attend its meetings to give special technical advice. Such persons shall not have voting privileges, but may ask permission to have their opinion recorded in the minutes.

II. The continuance of the records of the ICRP shall be in the hands of a Secretary of the ICRP elected by the ICRP from among its regular members and subject to the approval of the IEC.

III. The ICRP shall familiarize itself with progress in the whole field of radiation protection. The Secretary shall be responsible for the preparation of a programme to be submitted to the Commission for discussion at its meetings. Preliminary reports shall be prepared and circularized to all members of the ICRP and other specially qualified individuals at least six months before the meeting of the Congress.

IV. The Chairman shall be elected by the ICRP during one Congress to serve through the succeeding Congress. The choice shall not be limited to the country in which it is proposed to hold the succeeding Congress.

V. Decisions of the ICRP shall be decided by a majority vote, with the Chairman casting the deciding vote in case of a tie. A minority opinion may be appended to the minutes of a meeting if so desired by any member and upon his submission of same in writing to the Secretary.

Policy of the Commission

(2) The policy adopted by the ICRP in preparing its recommendations is to deal with the basic principles of radiation protection, and to leave to the various national protection committees the right and the responsibility of introducing the detailed technical regulations, recommendations, or codes of practice best suited to the needs of their individual countries.

(3) The Commission's recommendations have been kept continually under review in order to cover the increasing number and scope of potential radiation hazards, and to amend safety factors in the light of new knowledge concerning the effects of ionizing radiations.

Official Relations with the World Health Organization

(4) In accordance with rules laid down by the World Health Assembly for the admission of non-governmental organizations into official relations with the World Health Organization, such relations were established between the ICRP and WHO in 1956 and were reaffirmed in 1958. This arrangement has been eminently satisfactory to the Commission and it is hoped that it will continue.

Composition of the ICRP and Its Committees

(5) During the preparation of these recommendations the ICRP has had the following composition:

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* Mr. Binks resigned as Secretary in 1957, for health reasons. After his resignation E. E. Smith (Great Britain) served as Acting Secretary, and since August 1, 1957, B. Lindell (Sweden) has served as Temporary Secretary.

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RECOMMENDATIONS OF THE COMMISSION

A. PREFATORY REVIEW

(1) Prior to the Geneva meeting of the Commission in April 1956, permissible levels of exposure to ionizing radiation had been expressed in terms of a dose in a rather short interval of time (1 day or 1 week), that is, essentially, in terms of an average dose rate—the average referring to the temporal distribution of the dose in the specified interval of time. Implicitly, if not explicitly, it was assumed that

* From March, 1958

if this average dose rate was low enough, no appreciable bodily injury would become apparent in the lifetime of the individual. The assumption was based largely on radiological experience which indicated that substantial skin recovery occurred within a few months following a moderate therapeutic dose and that the latent period for some long-term effects of radiation (e.g. cancer of the skin) resulting from residual tissue damage, was longer the lower the dose (or dose rate in the case of chronic exposure). Thus, in an occupationally exposed individual a long-term effect might not become apparent in his lifetime, even if a certain amount of permanent injury had occurred.

(2) The basic permissible weekly dose at that time was 0.3 rem/week. Assuming that a person was occupationally exposed at this rate (50 weeks a year) for 50 years, the permissible accumulated dose would be 750 rems in the most critical organs or essentially throughout the body. It was realized then that this constituted a "large" lifetime dose and an appropriate warning was included in the Commission's report of 1955.

(3) The general awareness of radiation hazards, induced caution on the part of those responsible for the protection of workers. Administratively, liberal factors of safety were often used especially in large atomic energy installations. As a result it was found that in general the actual exposure of personnel was kept at levels much below the then existing permissible limits.

(4) At the 1956 meeting of the Commission it became evident that stricter recommendations were needed. The 1955 Conference on the Peaceful Uses of Atomic Energy had aroused great interest in the development of atomic power plants throughout the world. In time this would greatly increase the number of persons occupationally exposed and would also bring about actual or potential exposure of other persons and the population as a whole. More importantly, the pressure for producing power economically might well do away with the "safety factors" mentioned above. Also, the average duration of occupational exposure per individual worker might increase. On the biological side it was considered that perhaps "recovery" plays a less important part in the long-term effects of radiation to be expected from continued exposure at low levels, than was earlier supposed. Because of the larger number of persons who would be exposed, occupationally or otherwise, genetic damage assumed greater importance. This was accentuated in no small degree by the realization that in some countries the per capita genetic dose contributed by medical procedures was about the same as that contributed by background radiation.

(5) Statistical studies had shown that the incidence of leukemia in radiologists was significantly greater than in other physicians who presumably were not professionally exposed to radiation. Of necessity these radiologists included those who had practiced their specialty at the time when radiation protection was not very effectively carried out. Therefore, the accumulated doses received by those who developed leukemia may have been much higher than the 750 rems mentioned above. On the other hand, since most of the exposure of these radiologists resulted from diagnostic procedures carried out with low voltage X-rays, the lifetime dose in the blood-forming organs may have been lower than 750 rems even if the skin dose, especially in some parts of the body was much higher. The mechanism of leukemia induction by radiation is not known. It may be postulated that if the dose is lower than a certain threshold value no leukemia is produced. In this case it would be necessary to estimate the threshold dose and to make allowances for recovery, if any. There is not sufficient information to do this, but caution would suggest that an accumulated dose of 750 rems might exceed the threshold. The most conservative approach would be to assume that there is no threshold and no recovery, in which case even low accumulated doses would induce leukemia in some susceptible individuals, and the incidence might be proportional to the accumulated dose. The same situation exists with respect to the induction of bone tumors by bone-seeking radioactive substances.

(6) Presently available longevity studies differ as to whether there is a statistically significant life shortening in radiologists as compared to other specialists presumably not occupationally

exposed to radiation. However, in mammals chronically exposed at different daily doses a definite effect on longevity becomes clearly apparent at the higher daily doses. If extrapolation to lower daily doses, and then to man, is justified, it may be concluded that occupational exposure at presently accepted permissible limits may entail some life shortening. This effect may be interpreted as a slight acceleration of the natural aging process.

(7) The effects just discussed illustrate the two different types of possible long-term somatic effect that must be considered in setting up permissible limits of exposure. The first type (leukemia) is a serious effect occurring in some individuals and, therefore, the aim of protection would be to reduce the incidence to the lowest practical limit. The second type (life shortening) is presumably an effect on every individual and, therefore, the aim of protection would be to reduce the degree of effect to the lowest practical value. The definition of permissible dose has been changed to include explicitly these two types of possible effect.

(8) Genetic effects manifest themselves in the descendants of exposed individuals. The injury, when it appears, may be of any degree of severity from inconspicuous to lethal. A slight injury will tend to occur in the descendants for many generations, whereas a severe injury will be eliminated rapidly through the early death of the individual carrying the defective gene. Thus the sum total of the effect caused by a defective gene until it is eliminated may be considered to be roughly the same. The main consideration in the control of genetic damage (apart from aspects of individual misfortune) is the burden to society in future generations imposed by an increase in the proportion of individuals with deleterious mutations. From this point of view it is immaterial in the long run whether the defective genes are introduced into the general pool by a few individuals who have received large doses of radiation, or by many individuals in whom smaller doses have produced correspondingly fewer mutations. However, even in this case it is desirable to limit the dose received by an individual.

(9) In view of the foregoing, recommendations are made in this report in terms of maximum permissible doses for individuals and for population groups. In either case limits are set on the basis of dose accumulated over a period of years rather than in terms of a weekly dose that could be received for an indefinite period of time. The concept of limiting the accumulated dose was introduced by the Commission at its 1956 meeting in Geneva. The limitation of accumulated dose suggested at the time corresponds roughly to a three-fold reduction in weekly dose, for example, in the case of whole body occupational exposure when the exposure takes place approximately at a constant rate.

(10) In practice the problem of chief concern is chronic exposure either at low dose rates or by intermittent small doses. Under these conditions it is reasonable to assume that the dose accumulated over a period of years is the controlling factor, *provided* the intermittent doses are sufficiently small. Thus, in addition to limiting the accumulated dose it is necessary to limit the magnitude of a single dose (that is, a dose received in a short interval of time). Previously a single exposure equal to the maximum permissible weekly dose ("seven consecutive days") was permitted. Following the same pattern, the single dose limit for occupational exposure recommended in the present report is expressed in terms of the maximum permissible dose accumulated in a period of "13 consecutive weeks". The recommended value for the relevant organ (e.g. 3 rems for the blood-forming organs) has been made as high as it appears prudent, in the light of present knowledge. The stipulation of any 13 *consecutive* weeks has been made to make sure that operations are carried out in such a way that intermittent doses approximating the full 13 week quota do not occur at short intervals.

(11) In the recommendations published in 1955 maximum permissible limits were set on the basis of doses received by certain organs and certain serious late effects known to occur in them with sufficiently large doses. Provisions were made by means of an arbitrary "dose distribution curve" (in the report of Committee I) to limit the dose in other organs and tissues. This was made necessary by the adoption of a

maximum permissible dose for the skin twice as large as that for the blood-forming organs (with an assumed effective depth of 5 cm). In the present report separate recommendations are made for three groups of organs or tissues:

(a) Blood-forming organs, gonads and lenses of the eyes.
(b) Skin and thyroid gland.
(c) All other organs or tissues, specifically as regards exposure essentially limited to the organ or tissue in question.

(12) For the blood-forming organs, gonads and the lenses of the eyes the limits for occupational exposure are set in terms of the dose accumulated at various ages, according to the formula $D = 5(N - 18)$, where D is the dose in rems and N is the age in years, with the additional stipulation that the dose accumulated during any 13 consecutive weeks shall not exceed 3 rems.

(13) For the skin and the thyroid gland the limit for occupational exposure is set in terms of the dose accumulated during any 13 consecutive weeks, and the recommended value is 8 rems. This is derived from an average of 0.6 rem/week (the maximum permissible weekly dose formerly recommended for the skin of the whole body) which in 13 weeks amounts to 7.8 rems, and the nearest whole number is used to avoid the implication of greater accuracy than is warranted by present knowledge. The limit for the dose in these tissues accumulated in 1 year is $(0.6 \times 50) = 30$ rems. It should be noted that the new recommendation refers to the dose in the skin itself, irrespective of the dose distribution in the subcutaneous tissues. Therefore, the comparison should be made with the previous recommendation for exposure to radiation of very low penetrating power, for which the recommended limit was 1.5 rem/week. Accordingly, in this case also a reduction has been made in the accumulated dose, but the single exposure limit has been increased from 1.5 to 8 rems. This should provide more flexibility in practice than was possible formerly.

(14) For all organs and tissues of the body except the blood-forming organs, the gonads and the lenses of the eyes, the limit for occupational exposure is set in terms of the dose accumulated during any 13 consecutive weeks. With the exception of the skin, the pertinent practical cases in this category relate to exposure from internal sources essentially limited to individual organs or tissues. The following points require consideration. Whereas in the case of the blood-forming organs, the gonads, the lenses of the eyes and the skin, the objective of protection is to prevent or minimize definitely known types of injury, in the case of other organs the type of injury is not known. (Bone constitutes the only exception, in which case the relevant injury is cancer and permissible limits may be set on the basis of data furnished by human subjects who accumulated radium in their skeletons.) Possibly, radiation in sufficient dosage may increase the incidence of cancer in one of these organs (e.g. the thyroid gland) or it may accelerate aging of the organ. In the absence of factual data, it was deemed prudent in earlier recommendations of the commission to set the maximum permissible limit for these organs, when irradiated by internal sources, as low as that for the more sensitive organs such as the gonads, that is, 0.3 rem/week. When the exposure is essentially limited to *one organ* because of the more or less selective accumulation of a certain radioactive isotope therein, it is obvious that this limit embodies a factor of safety not present when the *whole body* is exposed at the same permissible limit. For this reason and the fact that none of these organs is known to be as sensitive as the blood-forming organs, the gonads and the lenses of the eyes, the Commission has decided to retain the previously recommended maximum permissible dose of 0.3 rem/week for each organ singly (with some exceptions noted in the report of Committee II). However, the limit is now expressed in terms of 13 consecutive weeks, which makes it 4 rems, in round figures, with an annual accumulated dose of 15 rems. Committee II has made the necessary adjustments to conform with the lower permissible limits now recommended for some organs and for what may be regarded as constituting "whole body" exposure (e.g. isotopes distributed throughout the body, or several isotopes present simultaneously, each concentrating significantly in a different organ).

(15) The Commission has given particular

attention to the difficult problem of setting permissible limits for exposure of persons in the neighborhood of radiation installations. The chief obstacle is the almost complete lack of knowledge of the deleterious effects that may result from low level exposure starting at conception and continuing throughout life. It is reasonable to expect a more marked effect than in the case of exposure starting after the individual has reached maturity (for one thing, because the period of exposure is longer), but it is very difficult to decide what allowance to make. Guidance could be obtained from suitable experiments carried out with mammals and it is hoped that such studies will be undertaken soon in some laboratories. In the meantime caution is in order. The Commission recommends that provisions be made in a controlled area or areas to make sure that under normal operating conditions no child residing outside such controlled areas, receive more than 0.5 rem/year (in the appropriate organs) from radiation or radioactive material originating in the controlled area or areas. In practice it may be expected that while fluctuations in exposure rate would occur, they would not be such as to require special limitations. It will be noted that this is one tenth of the *lowest* annual dose in any organ permitted for occupational exposure. It includes contributions made by external and internal sources but does not include doses contributed by natural background radiation or medical procedures.

(16) Special groups of *adults* in the vicinity of a controlled area are permitted to receive larger annual doses in the gonads, the blood-forming organs and the lenses of the eyes, by a factor of three (i.e. 1.5 rems). No biological significance should be attached to the magnitude of this factor, since present radiobiological information is grossly inadequate in this respect. The value recommended (1.5 rems/year) is one tenth of the former maximum permissible annual dose for occupational exposure, on the basis of 0.3 rem/week in the most sensitive organs. (See also paragraphs 54, 56 and 57.)

(17) Planning for the future expansion of nuclear energy programs and the more extensive uses of radiation, requires measures intended to protect whole populations. Genetic damage is of greatest concern in this regard. The problem has been discussed by various national and international groups and tentative suggestions have been made. The Commission considered the problem at its 1956 meeting and later issued a statement in general terms. However, recommendations in quantitative terms are needed in the design of power plants and other radiation installations and particularly in making plans for disposal of radioactive waste products. It is of the utmost importance in this connection to make sure that nothing is done now that may prove to be a serious hazard later, which cannot be corrected at all or will be very expensive to correct. The Commission is aware of the fact that a proper balance between risks and benefits cannot yet be made, since it requires a more quantitative appraisal of both the probable biological damage and the probable benefits than is presently possible. Furthermore, it must be realized that the factors influencing the balancing of risks and benefits will vary from country to country and that the final decision rests with each country (insofar as operations within one country do not affect other countries).

(18) The Commission wishes to point out that it is important to assign quotas of a maximum permissible genetic dose to the different modes of exposure, in order to make sure that those responsible for the control of exposure in one category do not take up a disproportionate share of the permissible total in their planning. However, at this time it is deemed best not to assign rigid quotas. As a tentative guide an illustrative apportionment is appended to paragraph 65.

(19) Briefly, the suggested limit for the genetic dose was arrived at in the following manner: Estimates made by different national and international scientific bodies indicate that a per capita gonad dose of 6–10 rems accumulated from conception to age 30 from all man-made sources, would impose a considerable burden on society due to genetic damage, but that this additional burden may be regarded as tolerable and justifiable in view of the benefits that may be expected to accrue from the expansion of the practical applications of "atomic energy". There is at present considerable uncertainty as to the magnitude of

the burden (see for example the report of the United Nations Scientific Committee on the Effects of Atomic Radiation) and, therefore, it is highly desirable to keep the exposure of large populations at as low a level as practicable, with due regard to the necessity of providing additional sources of energy to meet the demands of modern society. A genetic dose of 10 rems from all man-made sources is regarded by most geneticists as the absolute maximum and all would prefer a lower dose. In some countries the genetic dose from medical procedures has been estimated to be about 4.5 rems (see *Report of Joint Study Made by ICRP-ICRU for the U.N. Scientific Committee*). Therefore, if the limit for the genetic dose from all man-made sources were set at 6 rems, the contribution from all sources other than medical procedures, would be limited to 1.5 rems in these countries. This would impose unacceptable restrictions on these countries. Accordingly, as a matter of practical necessity the Commission recommends that medical exposure be considered separately and that it be kept as low as is consistent with the necessary requirements of modern medical practice. The joint study of ICRP-ICRU indicates that careful attention to the protection of the gonads would result in a considerable reduction of the genetic dose due to medical procedures without impairment of their value. In view of these considerations the Commission suggests a limit of 5 rems for the genetic dose from all man-made sources of radiation and activities, except medical procedures.

(20) At the present time the contribution to the genetic dose from all man-made sources (other than medical procedures) is small. With careful planning the rate of increase can be kept under control and the ultimate value of this contribution may never reach the suggested limit of 5 rems. Since the genetic dose from medical exposure in most countries is much lower than 4.5 rems and since in those countries in which it is high efforts are being made to reduce it, the total genetic dose from all man-made sources actually received by the world population may be expected to be considerably less than 10 rems, perhaps even less than 6 rems in the foreseeable future. Furthermore, if a thermonuclear reaction can be utilized as a source of power, the problem of radiation protection may be greatly simplified.

(21) The Commission is aware that compliance with the new recommendations may entail structural changes in some existing installations and/or changes in operative procedures. Since in fact the new recommendations are more restrictive because of the greater emphasis put on the dose accumulated over a long period of time, it is not essential that such changes be made immediately, although it is obviously desirable. As a practical guide it is suggested that the transition period during which the necessary changes would be made, should not exceed five years.

(22) The Commission wishes to point out again that the setting up of maximum permissible limits of occupational and non-occupational exposure (especially the latter) requires quantitative information not yet available about the risks and benefits of an expanded use of "atomic energy". For this reason the Commission will be glad to receive factual data and suggestions from those concerned with the production or utilization of ionizing radiation, so that as much pertinent information as possible may be available to it in its future deliberations.

B. BASIC CONCEPTS

Objectives of Radiation Protection

(23) Exposure to ionizing radiation can result in injuries that manifest themselves in the exposed individual and in his descendants: these are called somatic and genetic injuries respectively.

(24) Late somatic injuries include leukemia and other malignant diseases, impaired fertility, cataracts and shortening of life. Genetic injuries manifest themselves in the offspring of irradiated individuals, and may not be apparent for many generations. Their detrimental effect can spread throughout a population by mating of exposed individuals with other members of the population.

(25) The objectives of radiation protection are to prevent or minimize somatic injuries and to minimize the deterioration of the genetic constitution of the population.

Critical Organs and Tissues

(26) The organs and tissues of the body exhibit varying degrees of radiosensitivity, and it is therefore necessary, for purposes of protection, to consider their radiosensitivity with respect to specific functions as well as the doses they receive. When this is done, some organs and tissues assume a greater importance, according to the circumstances under which they are irradiated. They are then said to be critical.

(27) In the case of more or less uniform irradiation of the *whole body*, the critical tissues are those tissues of the body that are most radiosensitive with respect to the ability of carrying out functions essential to the body as a whole. In this report these are taken to be the blood-forming organs, the gonads, and the lenses of the eyes. In previous reports the skin was listed as a critical organ in the case of whole body exposure. The presentation of the recommendations in the present report is simplified by not designating the skin as a critical organ.

(28) In the case of irradiation more or less limited to *portions of the* body, the critical tissue is that tissue most likely to be permanently damaged either because of its inherent radiosensitivity, or because of a combination of radiosensitivity and localized high dose.

Permissible Dose

(29) Any departure from the environmental conditions in which man has evolved may entail a risk of deleterious effects. It is therefore assumed that long continued exposure to ionizing radiation additional to that due to natural radiation involves some risk. However, man cannot entirely dispense with the use of ionizing radiations, and therefore the problem in practice is to limit the radiation dose to that which involves a risk that is not unacceptable to the individual and to the population at large. This is called a "permissible dose"

(30) The permissible dose for an *individual* is that dose, accumulated over a long period of time or resulting from a single exposure, which, in the light of present knowledge, carries a negligible probability of severe somatic or genetic injuries; furthermore, it is such a dose that any effects that ensue more frequently are limited to those of a minor nature that would not be considered unacceptable by the exposed individual and by competent medical authorities.

(31) Any severe somatic injuries (e.g. leukemia) that might result from exposure of individuals to the permissible dose would be limited to an exceedingly small fraction of the exposed group; effects such as shortening of life span, which might be expected to occur more frequently, would be very slight and would likely be hidden by normal biological variations. The permissible doses can therefore be expected to produce effects that could be detectable only by statistical methods applied to large groups.

(32) The permissible dose to the gonads for the *whole population* is limited primarily by considerations with respect to genetic effects (see paragraphs 58–65).

Categories of Exposure

(33) These recommendations are designed to limit not only somatic but also genetic effects; it is therefore necessary to reduce as much as possible the dose to the population as a whole, as well as to the individual. In general, doses resulting from all sources of ionizing radiation should be considered in the appraisal of possible biological damage. However practical considerations make it necessary to consider separately the doses resulting from two categories of exposure, namely:

(a) Exposure to natural background radiation.
(b) Exposure resulting from medical procedures.

(34) Natural background radiation varies considerably from locality to locality and the doses it contributes to the various organs are not well known. If maximum permissible limits recommended by the Commission included background radiation, the allowable contribution from man-made sources—which are the only ones that can be controlled—would be uncertain and would have to be different for different localities. Accordingly, doses resulting

from natural background radiation are excluded from all maximum permissible doses recommended in this report.

(35) In medical procedures, exposure of the patient to primary radiation is generally limited to parts of the body, but the whole body is exposed to some extent to stray radiation. The contributions to the doses in various organs and the part played in the over-all effects on the individual are practically impossible to evaluate at the present time. The Commission recognizes especially the importance of the *gonad* doses resulting from medical exposure and the attendant genetic hazard to the population. Accordingly, it recommends that the medical profession exercise great care in the use of ionizing radiation in order that the gonad dose received by individuals before the end of their reproductive periods be kept at the minimum value consistent with medical requirements. However, individual doses resulting from medical exposure are excluded from all maximum permissible doses recommended in this report.

(36) The recommendations cover the following categories of exposure. In principle both the exposure of *individuals* and averages over the whole *population* have to be considered, but recommendations with regard to individual exposure are given only for the groups A and B.

A. *Occupational exposure.*
B. *Exposure of special groups:*
   (a) Adults who work in the vicinity of controlled areas (see paragraphs 71 and 72), but who are not themselves employed on work causing exposure to radiation.
   (b) Adults who enter controlled areas occasionally in the course of their duties, but are not regarded as radiation workers.
   (c) Members of the public living in the neighborhood of controlled areas.
C. *Exposure of the population at large.*
D. *Medical exposure.*

Occupational exposure

(37) Exposure of an *individual* who normally works in a controlled area constitutes occupational exposure. Maximum permissible doses are set for the individuals in the small portion of the population that can be occupationally exposed (paragraphs 46–52). The contribution from this group to the genetic dose to the *population* as a whole is discussed in paragraph 65.

Exposure of special groups

(38) Persons who only occasionally enter a controlled area and persons who work or reside in the vicinity of a controlled area may be exposed to radiation originating in the controlled area. They constitute groups that may include children and pregnant women as well as individuals subject to other hazards, and may in total constitute a large fraction of the whole population. For these reasons the maximum permissible dose to these persons as *individuals* is set lower than for persons occupationally exposed (paragraphs 53–57). The contribution from these groups to the genetic dose to the whole *population* is discussed in paragraph 65.

Exposure of the population at large

(39) Members of the population at large may be exposed to radiation that cannot be related to any specific controlled area; e.g. exposure from environmental contamination and widely distributed radiation sources such as wristwatches, TV-sets and various applications of radioactive materials to be expected as a result of future expansion in the atomic energy field. As such exposure is not easily controlled, it will be impossible to ensure that a recommended maximum permissible individual dose is not exceeded in any single case. Where large numbers are involved, it will not be possible to examine the habits of every individual. A reasonable procedure would be to study a sample of the group involved and to set the environmental level so that no individual in the sample receives any excessive exposure. There will always remain the possibility that someone of grossly different habits from those in the observed sample may receive a higher dose than the maximum in the sample.

(40) In order to facilitate planning for the anticipated increased uses of nuclear energy and other sources of radiation, it is desirable at this time to recommend a maximum for the genetic dose to the *population* (paragraph 64); this

maximum will determine what average gonad exposure could be allowed. Part of the recommended maximum genetic dose will have to be used for exposure of groups such as A and B and for medical exposure. The proper apportionment for exposure of the population at large must allow for both internal and external exposure (paragraph 65).

Medical exposure

(41) No recommendations are given with regard to the dose to the individual from medical exposure. (The contribution of medical exposure to the genetic dose is discussed in paragraphs 69–70.)

Reduction in Maximum Permissible Dose

(42) The new recommendations were introduced partly with the intention of limiting the genetically significant radiation exposure (see paragraph 63) of the population, and partly to limit the probability of somatic injury by reducing the lifetime dose. This reduction does not result from positive evidence of damage due to the use of the earlier permissible dose levels, but rather is based on the concept that biological recovery may be minimal at such low dose levels.

Time Interval over which Dose is to be Assessed

(43) The maximum permissible weekly doses recommended by the Commission in 1950 have been replaced by limits for the doses received over longer periods of time (paragraphs 47–49). In the case of occupational exposure the maximum permissible dose that may be accumulated at a certain time depends on the age of the worker. The dose to individuals in the population at large, or in special groups other than occupational, may be accumulated at a rate that is determined by a maximum permissible annual dose. The genetic dose to the whole population is assessed over the period between conception of the individual and conception of each child of the individual. (See paragraph 63 for method of evaluation.)

(44) These extended periods of time allow for some flexibility in the way in which radiation exposure may be received, and at the same time provide what is considered to be adequate protection for each group of the population.

C. MAXIMUM PERMISSIBLE DOSES

GENERAL

(45) It is emphasized that the maximum permissible doses recommended in this section are *maximum* values; the Commission recommends that all doses be kept as low as practicable, and that any unnecessary exposure be avoided.

EXPOSURE OF INDIVIDUALS

Occupational Exposure

(46) In any organ or tissue, the *total* dose due to occupational exposure shall comprise the dose contributed by external sources during working hours and the dose contributed by internal sources taken into the body during working hours. It shall not include any medical exposure or exposure to natural radiation.

Exposure of the gonads, the blood-forming organs and the lenses of the eyes

(47) The maximum permissible total dose accumulated in the gonads, the blood-forming organs and lenses of the eyes at any age over 18 years shall be governed by the relation

$$D = 5(N - 18)$$

where D is tissue dose in rems and N is age in years.

(48) For a person who is occupationally exposed at a constant rate from age 18 years, the formula implies a maximum weekly dose of 0.1 rem. It is recommended that this value be used for purposes of planning and design.

Rate of dose accumulation

(49) To the extent the formula permits, an occupationally exposed person may accumulate the maximum permissible dose at a rate not in excess of 3 rems during any period of 13 consecutive weeks (i.e. in no 13 consecutive weeks shall the dose exceed 3 rems). If necessary, the 3 rems may be received as a single dose, but as the scientific knowledge of the biological effects of differing dose rates is scant, single doses of the order of 3 rems should be avoided as far as practicable.

Application to special cases

(50) Setting permissible limits of exposure in terms of the dose accumulated up to a given age introduces certain practical complications. Thus, some workers (previously exposed at levels within the then permissible limits) may have already accumulated a dose in excess of the maximum permitted by the formula. There are also special cases in which exceptions in the application of the formula may be desirable for practical reasons and are justifiable within the context of paragraph 42. The following recommendations are intended to provide guidance on administrative policy, which may well vary according to circumstances at the local level. (It should be noted that this situation will obtain only during a relatively short transition period.)

(51a) *Previous exposure history unknown.* When the previous occupational exposure history of an individual is not definitely known, it shall be assumed that he has already received the full quota permitted by the formula.

(51b) *Persons exposed in accordance with the former maximum permissible weekly dose.* Persons who were exposed in accordance with the former maximum permissible weekly dose of 0.3 rem and who have accumulated a dose higher than that permitted by the formula, should not be exposed at a rate higher than 5 rems in any one year, until the accumulated dose at a subsequent time is lower than that permitted by the formula.

(51c) *Persons starting work at an age of less than 18 years.* When a person begins to be occupationally exposed at an age of less than 18 years, the dose shall not exceed 5 rems in any one year under age 18, and the dose accumulated to age 30 shall not exceed 60 rems. (The minimum age at which occupational exposure is legally permitted is lower than 18 years in some countries.)

(51d) *Accidental high exposure.* An accidental high exposure that occurs *only once in a lifetime* and contributes no more than 25 rems shall be added to the occupational dose accumulated up to the time of the accident. If the sum then exceeds the maximum value permitted by the formula, the excess need not be included in future calculations of the person's accumulated dose. Accidental exposure to doses higher than 25 rems must be regarded as being potentially serious, and shall be referred to competent medical authorities for appropriate remedial action and recommendations on subsequent occupational exposure. This is intended as an administrative guide to permit the continuation of work with radiation, following a bona fide accident ("once in a lifetime"), in cases in which interruption of such work, or curtailment of exposure, would handicap the individual in the pursuit of his career.

(51e) *Emergency exposure.* Emergency work involving exposure above permissible limits shall be planned on the basis that the individual will not receive a dose in excess of 12 rems. This shall be added to the occupational dose accumulated up to the time of the emergency exposure. If the sum then exceeds the maximum value permitted by the formula, the excess shall be made up by lowering the subsequent exposure rate so that within a period not exceeding 5 years, the accumulated dose will conform with the limit set by the formula. Women of reproductive age shall not be subjected to such emergency exposure.

Exposure of single organs other than the gonads, the blood-forming organs and the lenses of the eyes

(52) For exposure that is essentially restricted to portions or single organs of the body, with the exception of the gonads, the blood-forming organs and the lenses of the eyes, a higher dose than the one derived from the formula $D = 5(N - 18)$ is permitted. The following recommendations are made.

(52a) *A maximum dose of 8 rems/13 weeks for*

*the skin.** The earlier recommended weekly maximum doses for exposure of the skin, namely 0.6 rem in connection with a whole body exposure and 1.5 rems for exposure with radiation of very low penetrating power, have been replaced by the recommendation that the dose in the skin, accumulated over any 13 consecutive weeks, shall not exceed 8 rems. This recommendation now applies to all exposure of the skin, except the skin of the hands and forearms, feet and ankles. As the 8 rems are derived from an average of 0.6 rem/week, the annual dose for a 50 week year is limited to 30 rems.

(52b) *A maximum dose of* 20 *rems*/13 *weeks for the hands and forearms, feet and ankles.* The earlier recommended weekly maximum of 1.5 rems for exposure of hands and forearms, feet and ankles has been replaced by the recommendation that the dose accumulated over any 13 consecutive weeks shall not exceed 20 rems. This recommendation applies to all tissues of the above-mentioned extremities. As the 20 rems are derived from an average of 1.5 rems/week, the annual dose for a 50 week year is limited to 75 rems.

(52c) *A maximum dose of* 4 *rems*/13 *weeks for limited exposure of internal organs other than the thyroid, the gonads and the blood-forming organs.* In the case of internal organs, limited exposure originates almost exclusively from radioisotopes within the body. As most planning of release of radioactive isotopes to the air and water supplies in controlled areas is made under conditions where occupational groups rather than single individuals are exposed, a formula corresponding to the one given in paragraph 47 cannot in general be applied to internal exposure. An average of 0.3 rem/week in the organ of interest (with some exceptions noted in the report of Committee II) is expected to be maintained under equilibrium conditions if the concentration in air *or* water of the relevant isotope is kept at levels given in the tables in the report of Committee II. Variations of the dose rate will occur in practice, and are permissible, provided that the dose accumulated over any 13 consecutive weeks does not exceed 4 rems. As this maximum is derived from a weekly average of 0.3 rem, the annual dose for a 50 week year is limited to 15 rems.

(52d) *Whole body exposure from uptake of several radioisotopes.* When the radioactive isotopes in a mixture are taken up by several organs and the resultant tissue doses in such organs are of comparable magnitude, the combined exposure is considered to constitute essentially whole body exposure. Accordingly, the permissible levels of exposure will be those applicable to the gonads, the blood-forming organs and the lenses of the eyes.

Exposure of Special Groups

(53) The total maximum permissible individual dose shall consist of the sum of the doses contributed by both external and internal sources. It shall not include any medical exposure or exposure to natural radiation.

Total annual dose to the gonads, the blood-forming organs and the lenses of the eyes

(54) For any individual in the groups B(a) and B(b) (see paragraph 36), the total annual dose, including contributions from external and internal sources, to the gonads, the blood-forming organs and the lenses of the eyes, due to operations within the controlled area shall not exceed 1.5 rems, nor shall the contribution from a mixture of isotopes whose combined exposure constitutes essentially whole body exposure, make the total annual dose exceed 1.5 rems. There is an exception in the case of the skin and the thyroid where an annual maximum of 3 rems is allowed.

(55) The group B(c) differs from the groups B(a) and B(b) in that it contains children for whom it is considered that a lower figure, namely 0.5 rem/year (in the gonads, the blood-forming organs and the lenses of the eyes), should apply. Accordingly the presence of children in the group B(c) will require the use of a value of 0.5 rem/year for this group for purposes of planning and design.

Internal exposure of single organs

(56) The *individual* maximum permissible annual dose will not be exceeded *from internal exposure* of any single organ, if the release of radioactive material is planned on the basis

* This also applies to the thyroid, see report of Committee II.

of $\frac{1}{10}$ of the maximum permissible concentration (MPC) in air *or* water as given for continuous occupational exposure ("168-hour week") in the tables in the report of Committee II.

Whole body exposure from uptake of several radioisotopes applicable to group B(c)

(57) When the radioactive isotopes in a mixture are taken up by several organs in such a way that the combined exposure constitutes essentially whole body exposure, a reduction of the maximum permissible concentrations based on the exposure of single organs (see report of Committee II) becomes necessary. The reduction should take into account the number and importance of the organs in question and the contribution to whole body exposure made by radioactive material in the circulating blood and in organs other than those in which it is assumed to concentrate. In this case the total body is considered to be the critical organ and the MPC values for the individual radioisotopes are reduced so that the MPC for the mixture corresponds to an average annual dose of 0.5 rem to the total body. For such mixtures of radioisotopes this is equivalent to a reduction in the MPC values of the individual radioisotopes in the mixture by a factor of $\frac{1}{3}$ or less in addition to the factor of $\frac{1}{10}$ recommended in paragraph 56.

EXPOSURE OF POPULATION

General

(58) Proper planning for nuclear power programs and other peaceful uses of atomic energy on a large scale requires a limitation of the exposure of whole populations, partly by limiting the individual doses and partly by limiting the number of persons exposed.

(59) This limitation necessarily involves a compromise between deleterious effects and social benefits. Consideration of genetic effects plays a major role in its evaluation. The problem has been discussed extensively in recent years and suggestions have been made by different national bodies. The Commission is aware of the fact that a proper balance between risks and benefits cannot yet be made, since it requires a more quantitative appraisal of the probable biological damage and the probable benefits than is presently possible. Furthermore, it must be realized that the factors influencing the balancing of risks and benefits will vary from country to country and that the final decision rests with each country.

(60) Because of the urgent need for guidance in this regard, the Commission in the following sections suggests an interim ceiling for the exposure of the whole population. The proposed level is essentially in accordance with suggestions by other scientific groups that have studied the problem and tried to evaluate the possible biological effects. It is felt that this level provides reasonable latitude for the expansion of atomic energy programs in the foreseeable future. It should be emphasized that the limit may not in fact represent the proper balance between possible harm and probable benefit, for reasons already mentioned.

(61) On the assumption that the genetic effects are linearly related to the gonad dose and provided that no threshold dose exists, it is possible to define a population dose average that is relevant to the assessment of genetic injury to the whole population (paragraphs 62–63). In the case of somatic effects no such dose can easily be defined although the annual per capita dose to certain tissues or to the whole body may be relevant on the assumption of a non-threshold, linear dose-effect relation.

Genetic Dose

Assessment of genetic dose

(62) The genetic dose to a population is the dose which, if it were received by each person from conception to the mean age of childbearing, would result in the same genetic burden to the whole population as do the actual doses received by the individuals. A permissible genetic dose is that dose, which if it were received by each person from conception to the mean age of childbearing, would result in an acceptable burden to the whole population.

(63) The genetic dose to a population can be assessed as the annual genetically significant dose multiplied by the mean age of childbearing, which for the purpose of these recommendations is taken to be 30 years. The annual genetically significant dose to a population is the average of the individual gonad doses, each weighted for the expected number of children conceived subsequent to the exposure.

Maximum permissible genetic dose

(64) It is suggested that the genetic dose (see paragraph 63) to the whole population from all sources additional to the natural background should not exceed 5 rems plus the lowest practicable contribution from medical exposure. The background is excluded from the suggested value because it varies considerably from country to country. The contribution from medical exposure is considered separately for the same reason and also because the subject is being studied for the purpose of limiting such exposure to the minimum value consistent with medical requirements.

Apportionment of genetic dose

(65) The suggested maximum genetic dose of 5 rems in addition to the dose from medical procedures and natural background must not be used up by one single type of exposure. The proper apportionment of the total will depend upon circumstances which may vary from country to country, and the decision should therefore be made by national authorities.

Addendum to paragraph 65

(a) *Illustrative apportionment.* The Commission does not wish to make any firm recommendations as to the apportionment of the genetic dose of 5 rems but, for guidance, gives the following apportionment as an illustration.

(A) Occupational exposure	1.0 rem
(B) Exposure of special groups	0.5 rem
(C) Exposure of the population at large	2.0 rems
Reserve	1.5 rems

(b) *Fractions of population.* Assuming that the ratio of the total population to the reproductive population is the same in all groups, the largest fraction (ε) of the whole population that can be exposed to an average annual dose $\bar{D}_1^i$ is given by the equation:

$$\varepsilon \cdot 30 \cdot \bar{D}_1^i = D_{30}^i$$

where D_{30}^i is the apportionment of the genetic dose to the ith exposure group, and the average annual dose within the group can be expressed as a fraction of the maximum permissible individual annual dose; i.e. $\bar{D}_1^i = F_1 D_1^i$.

(c) *Occupational exposure.* Assigning 1.0 rem to occupational exposure would mean that 0.7 per cent of the whole population could accumulate the maximum permissible occupational gonad dose of 60 rems by age 30. It is very unlikely that such a figure will be approached in the foreseeable future. At the present time, the number of persons occupationally exposed in technologically developed countries is about 0.1–0.2 per cent of the population, and most of these persons receive doses which are considerably less than the maximum permissible doses.

(d) *Exposure of special groups.* Since the contribution from the special groups is largely due to group B(c) an apportionment of 0.5 rem for the special groups would imply that about 3 per cent could be exposed to the maximum permissible individual annual gonad doses for these groups. The allowable size of these groups varies inversely with the average dose within the groups. Thus, if this dose were only 10 per cent of the maximum individual doses, the groups could amount to 30 per cent of the whole population, which is very much larger than is likely to occur.

(e) *Exposure of the population at large.* The apportionment of 2.0 rems (with a long-term reserve of 1.5 rems for possible eventualities) for the genetic exposure of the population at large is intended for planning purposes in the development of nuclear energy programs (with the associated waste disposal problems) and more extensive uses of radiation sources. In the case of internal exposure, the radioisotopes of interest are those that contribute to the gonad dose directly (by local concentration) or indirectly (by radiation produced elsewhere in the body). In either case the maximum permissible concentrations in air and water of these isotopes recommended by Committee II for continuous occupational exposure ("168-hour week") are based on an average yearly dose of 5 rems in the gonads or the whole body. If for these isotopes the average concentrations in public air and water supplies are lower than the values recommended for continuous occupational exposure by a factor of 1/100, the genetic dose to the population would amount to 1.5 rems (5 × 1/100 rems/year in the gonads ×

30 years = 1.5 rems). In this case the contribution from external sources should be limited to 0.5 rem in order not to exceed the total of 2 rems.

Somatic Dose

(66) No specific recommendations are made at this time as to the maximum permissible "somatically" relevant dose to the population. However, it is expected that the maximum permissible limits of the *individual* total doses recommended in paragraphs 46–57 will keep the average dose in any tissue at such a level that the injuries that could possibly occur in a population would be well within acceptable limits. (See paragraph 31.)

(67) In the case of external exposure of the whole body to penetrating radiation the restriction imposed by the genetic dose to the population automatically reduces the doses to internal organs below the *individual* maximum permissible annual doses recommended in paragraphs 46–57. Thus the probability of somatic injury in these organs is considerably lower than indicated in paragraph 66. The same thing applies to internal exposure resulting from radioisotopes that directly or indirectly contribute to the gonad dose of a population. (See addendum to paragraph 65.)

(68) There remain for further consideration those isotopes that concentrate in specific organs (other than the gonads). In view of the existing uncertainty as to the dose-effect relationships for somatic effects, it is suggested that for planning purposes the average concentrations of such isotopes, or mixtures thereof, in air or water, applicable to the population at large, should not exceed one-thirtieth of the MPC values for continuous occupational exposure given in the report of Committee II.

Medical Exposure

(69) Estimates of the current annual genetically significant dose from medical procedures in various countries were surveyed in a report of a joint study group of the ICRP and ICRU to the United Nations Scientific Committee on the Effects of Atomic Radiation. The United Nations Scientific Committee gives the following summary statement in its 1958 report to the General Assembly:

"The contribution made by medical procedures to the radiation exposure of populations has only lately been estimated and has increased very rapidly in some countries in recent years, so that it is difficult to evaluate such genetic and somatic effects as are associated with an increasing employment of radiological procedures in medicine. No information is yet available for prediction of the future trend of medical exposures. It is expected that improvements in equipment and techniques may considerably reduce individual exposures, but the ever-expanding use of X-rays may well increase the world population dose. Precautions of the type described by the International Commissions on Radiological Protection and on Radiological Units and Measurements should make possible such reduction of exposure to radiation as is without detriment to the medical value of these procedures."

(70) Reported values of the annual genetically significant dose in various countries range up to about 150 mrems, corresponding to a genetic dose of 4.5 rems (from conception to age 30 years, averaged over the whole population). Even with the highest values reported (corresponding to the genetic dose of 4.5 rems), the total genetic dose from all types of exposure would not exceed 10 rems if the apportionment for the contributions from other exposure sources than medical is not exceeded. There are indications that the highest levels of medical exposure reported could be reduced significantly by careful attention to techniques. On the other hand, the use of X-rays may increase, although probably to a lesser degree in the countries which now report the most extensive use. A certain allowance for medical exposure of populations must be made and may be made in the near future as the subject is being studied for the purpose of limiting such exposure to the minimum value consistent with medical requirements.

D. GENERAL PRINCIPLES REGARDING WORKING CONDITIONS

Responsibility

The owner or the person in charge of a controlled area shall be responsible for the working conditions and for the instruction of all persons working in the area regarding radiation hazards and methods of control. He shall be responsible for maintaining radiation levels outside the controlled area resulting from operations within the area so that exposures do not exceed the maximum permissible doses recommended in paragraphs 53–57.

(71) A controlled area shall be established where persons occupationally exposed could receive doses in excess of 1.5 rems/year.

(72) A controlled area is an area in which the exposure of personnel to radiation or radioactive material is under the supervision of a radiation protection officer.

(73) A radiation protection officer is one who has the knowledge and responsibility to apply appropriate radiation protection regulations. He may be the owner or the person in charge of the controlled area or he may be a technically competent person appointed by the above.

(74) A qualified expert (or health physicist) is a person having the knowledge and training needed to measure ionizing radiations and to advise regarding radiation protection. The qualification should be of the type specified by a National Committee.

Radiation Surveys and Monitoring

Surveys prior to use of controlled areas

(76) In those instances where the operations in a controlled area may disturb or alter significantly the environment with respect to radiation hazards, adequate surveys should be made of the radioactivity of the air, soil, and water prior to the start of operations. This will provide a base line from which to judge the adequacy of radiation controls within the area.

(77) During and after installation, appropriate radiation surveys shall be made to ensure that the pertinent recommendations will be complied with when routine operations commence. Routine operations shall be deferred until such compliance is assured.

(78) When additional operations are planned in the area, a thorough survey should be made of the background radiation prior to the start of the new operations. This will aid in the identification of the operation responsible for any increase of the background radiation or the contamination in the area.

Routine surveys and monitoring

(79) Radiation surveys shall be made regularly, at a frequency dictated by the operations within the area, to determine the adequacy of safety procedures. This should include checks of the facilities, equipment (radiation warning devices, radiation shields, hoods, respirators, ventilating system etc.), and working techniques. When there is any reasonable probability of a radiation hazard existing, the vicinity of the controlled area should also be surveyed.

Special surveys

(80) Specific and detailed recommendations regarding radiation surveys applicable to some special cases will be given in the Reports of the Committees of ICRP.

Health Surveillance

Pre-employment examination

(81) All new personnel in radiation work shall have a pre-employment medical examination. Notes should be made of the family history, of the previous occupational history, and of previous X-ray diagnostic examination or radiation therapy. The examination shall include a complete blood count, with determination of erythrocyte and leukocyte levels and a differential white cell count. It should be recognized that the examination is directed toward determining the "normal" condition of the worker at the time of employment, and toward noting any abnormalities that might later be confused with radiation damage.

(82) In cases where there has been previous occupational exposure, the total accumulated dose shall be recorded (see paragraphs 47–51) and any appropriate additional medical examinations performed. These should include

ophthalmological examinations, with particular reference to changes in the lens in cases of exposure to neutrons and to heavy particles, and examinations of skin and nails in the case of partial external irradiation and external contamination.

Routine medical examinations

(83) Medical examinations should be performed at a frequency depending upon the conditions of the occupational exposure. Blood counts, although they are a part of a medical examination, are not to be considered as a method of radiation monitoring.

(84) Persons occupationally exposed to neutrons and to heavy particles of significant penetration should have ophthalmological examinations, with particular reference to changes in the lens. The frequency of the examination will depend upon the conditions of exposure.

Personnel Monitoring

External radiation

(85) Doses received as a result of occupational exposures shall be systematically checked with appropriate instruments to ensure that the maximum permissible doses are not exceeded and to make it possible to keep individual cumulative dose records.

Internal radiation

(86) Tests should be performed to estimate the total body burden for workers who deal with unsealed radioactive isotopes that may give rise to levels of ingestion or inhalation in excess of the maximum permissible concentrations. Such tests should also be performed where radioisotopes may enter the body through the skin or through skin punctures and open wounds. These tests may require the monitoring of breath and excreta, and the direct determination of the body burden by means of a total body monitor, according to circumstances. The radiation doses delivered to the appropriate organs or tissues should be calculated and noted on the personal record, and the permitted doses of external radiation should be adjusted to allow for the "internal" doses.

Radiation Warning Devices

(87) An appropriate form of warning shall be provided to indicate the existence of a radiation hazard, even if the hazard is of a temporary nature only. In the latter case the warning device should be removed when the hazard no longer exists.

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ACCIDENTAL RADIATION EXCURSION AT THE OAK RIDGE Y-12 PLANT—II

HEALTH PHYSICS ASPECTS OF THE ACCIDENT*

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(*Received* 5 *November* 1958)

Abstract—The information presented here is one part of a series of four articles which collectively describe the accidental nuclear excursion which occurred June 16, 1958, in the Union Carbide Nuclear Company, Y-12 Plant at Oak Ridge, Tennessee. This discussion is submitted for publication in *Health Physics*, the official journal of the Health Physics Society in the interest of further dissemination of information concerning this unique event.

INTRODUCTION

On June 16, 1958, the first known accidental and unscheduled nuclear chain reaction to occur in an industrial process facility took place in the Union Carbide Nuclear Company Y-12 facility for the recovery of enriched uranium from fuel fabrication scrap and other salvage.

In an incident of this type, the hazard of first concern is not a high order nuclear explosion, but instead the lethal radiation which accompanies the incident. Of secondary importance, but of real concern to those who must combat the hazard, are the radiation after-effects. This discussion proposes to generally describe the conditions encountered during the course of the Y-12 incident and the health physics measures taken to evaluate and regain control of the area.

BACKGROUND

To best understand the Y-12 situation, distinction should be made between chemical processing facilities for the preparation of "cold" enriched uranium, which has relatively little radioactivity, and those designed to recover uranium from "hot" irradiated reactor fuel by separation from the highly radioactive fission products. In the latter case, protection must be routinely provided against the ever-present radiation hazard. Accordingly, such operations are shielded and controlled remotely. These requirements provide, in addition, a certain measure of protection against nuclear accident consequences. In the case of "cold" material, such as that handled in the Y-12 facility, the radiation problem is not acute; thus, the materials and equipment are accessible as a matter of routine. Personnel, in large numbers, frequent the area with relatively free access.

For many years, a successful program of nuclear safety has been carried out in Y-12. This program, while obligated to the principle of preventing even a single nuclear excursion was, and remains, contingent upon human capabilities. Since safety is never absolute, the program assumed that a nuclear excursion could occur. Accordingly, plans were made to provide capabilities to detect and combat such an incident. This plan may be described as follows.

1. *Detection of incident and sounding of alarm*

Although physical damage could occur, one cannot depend upon fuming, boiling, explosion, or a blue glow or haze as the only means for

* The information presented here was taken generously from material gathered and prepared through the unselfish efforts of many people, who contributed to the preparation of report Y-1234, "Accidental Radiation Excursion at the Y-12 Plant".

† Union Carbide Nuclear Company, a Division of Union Carbide Corporation Y-12 Plant. Contract No. W-7405-eng-26 with the United States Atomic Energy Commission.

detection of a nuclear reaction. Further, prompt action must be taken with a minimum of time lost to make decisions. Accordingly, γ-sensitive radiation detectors have been installed in all areas wherein potential criticality hazards exist. The monitor system in Building 9212, the location of the incident, is composed of six permanently-mounted instruments, a siren system audible throughout the building, and an annunciator system which indicates the location of an activated monitor. Any one of the six monitors automatically actuates the alarm sirens when the dose rate at the instrument exceeds 3 mr/hr. The utility of such a system is the fact that the alarm so given indicates the presence of a radiation hazard and implements emergency action with a minimum of lost motion. It cannot predict a nuclear excursion, and thus is after-the-fact, but it may, through the establishment of a prompt alert, minimize subsequent personnel exposures.

2. *Evacuation of area*

In case of a monitor alarm, quick orderly evacuation by the most direct route away from the processing area is considered to be the best path for the majority of people to follow. In some cases, it may be possible to provide sufficiently flexible evacuation signals so that evacuation routes are *always away* from the scene of a nuclear reaction. In the subject case, however, the former procedure is used due to the complexity of the plant layout, leaving to chance the possibility that any given evacuation path could pass closer to the accident location. By prompt evacuation of the affected area to preselected assembly stations, control over the group is established in case of need for further action.

3. *Initial survey of area and personnel*

β-γ survey meters are maintained in pairs at three locations at the perimeter of the area of concern. Local emergency squads, trained in survey procedures and maintained routinely in all hazardous areas, assemble immediately upon monitor alarm. They collect the instruments provided and survey the assembly areas, moving personnel as necessary to keep them in a radiation zone of less than 5 mr/hr. These teams also check for gross personal contamination ($>$1 r/hr) using the available portable survey instruments. Persons found to be contaminated should have their contaminated clothing removed immediately and should be sent to shower without delay.

4. *Screening of personnel for significant exposures*

Upon verification of the occurrence of a nuclear incident, evacuation of personnel proceeds to selected secondary locations where survey teams monitor badges for indium foil activation and check personal contamination further. As a matter of routine procedure, indium foils have been placed in all security badges for the purpose of quick identification of those persons who may have been exposed to neutron radiation arising from a nuclear reaction. The intent of this procedure is to identify those cases of significant exposure, particularly any which may be serious or critical. Such cases may then be directed to the Medical Dispensary for treatment and/or observation. Facilities for decontamination are available in these locations if needed.

5. *Follow-up area survey and rehabilitation*

As soon as feasible after activation of the plant emergency plan, trained survey teams, which are made available locally and through preestablished mutual assistance plans with others, are dispatched to the radioactive area. These teams monitor toward the reaction location and establish boundary lines at 12.5 mr/hr and 125 mr/hr. Simultaneously, efforts are made to determine the extent of environmental contamination by direct measurements and by air-sampling. Areas found to be unaffected, or within permissible limits, are released for rehabilitation as the surveys progress. Continuing control is exercised over the area until the radiation intensities permit access to the immediate area and suitable disposition is made of the source.

CONDITIONS ENCOUNTERED AND MEASURES TAKEN FOLLOWING THE INCIDENT

Preliminary surveys

At approximately 2.05 p.m., June 16, 1958, sirens of the radiation monitoring system

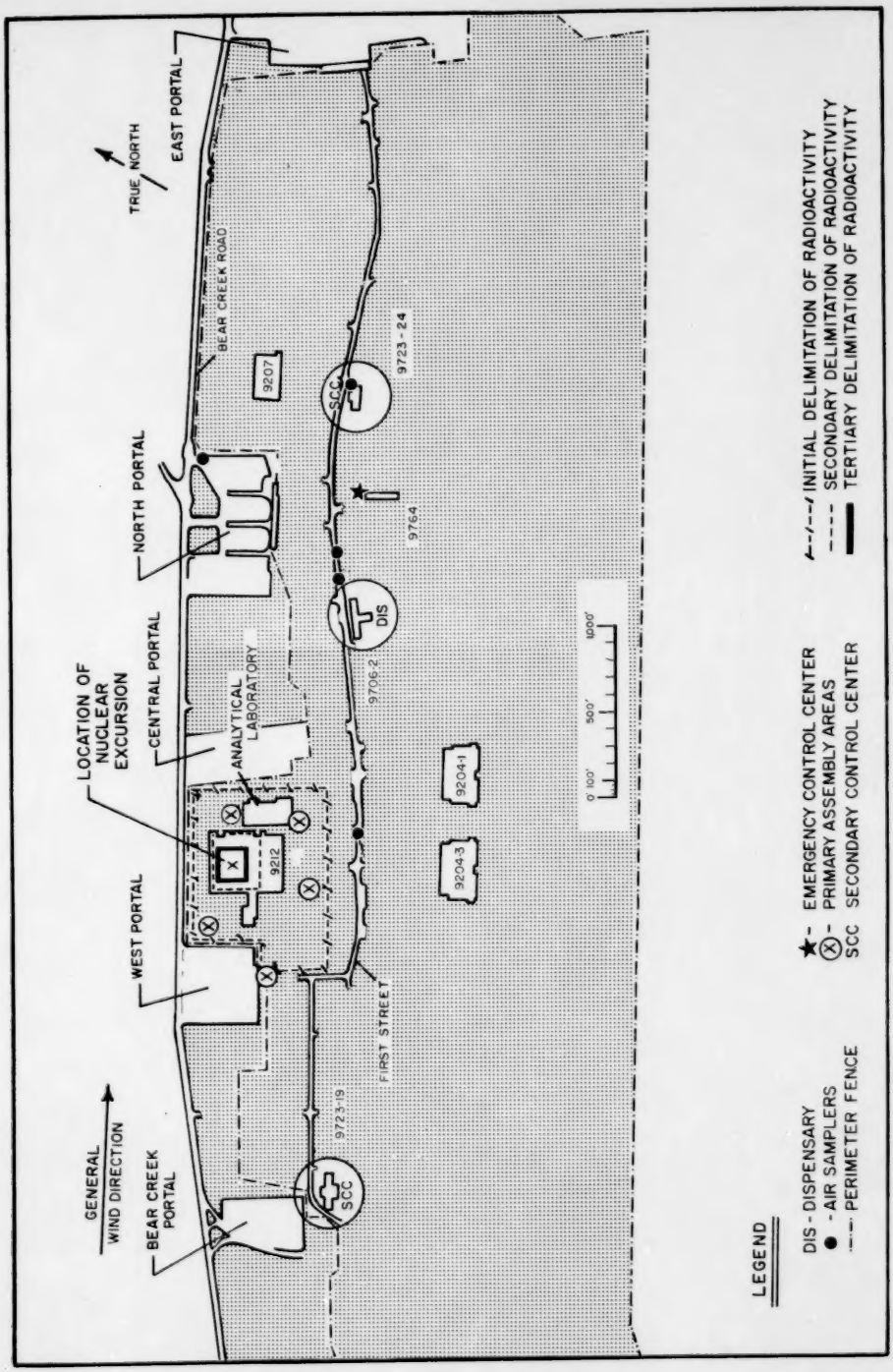


FIG. 1. Location of Y-12 Plant buildings pertinent to incident.

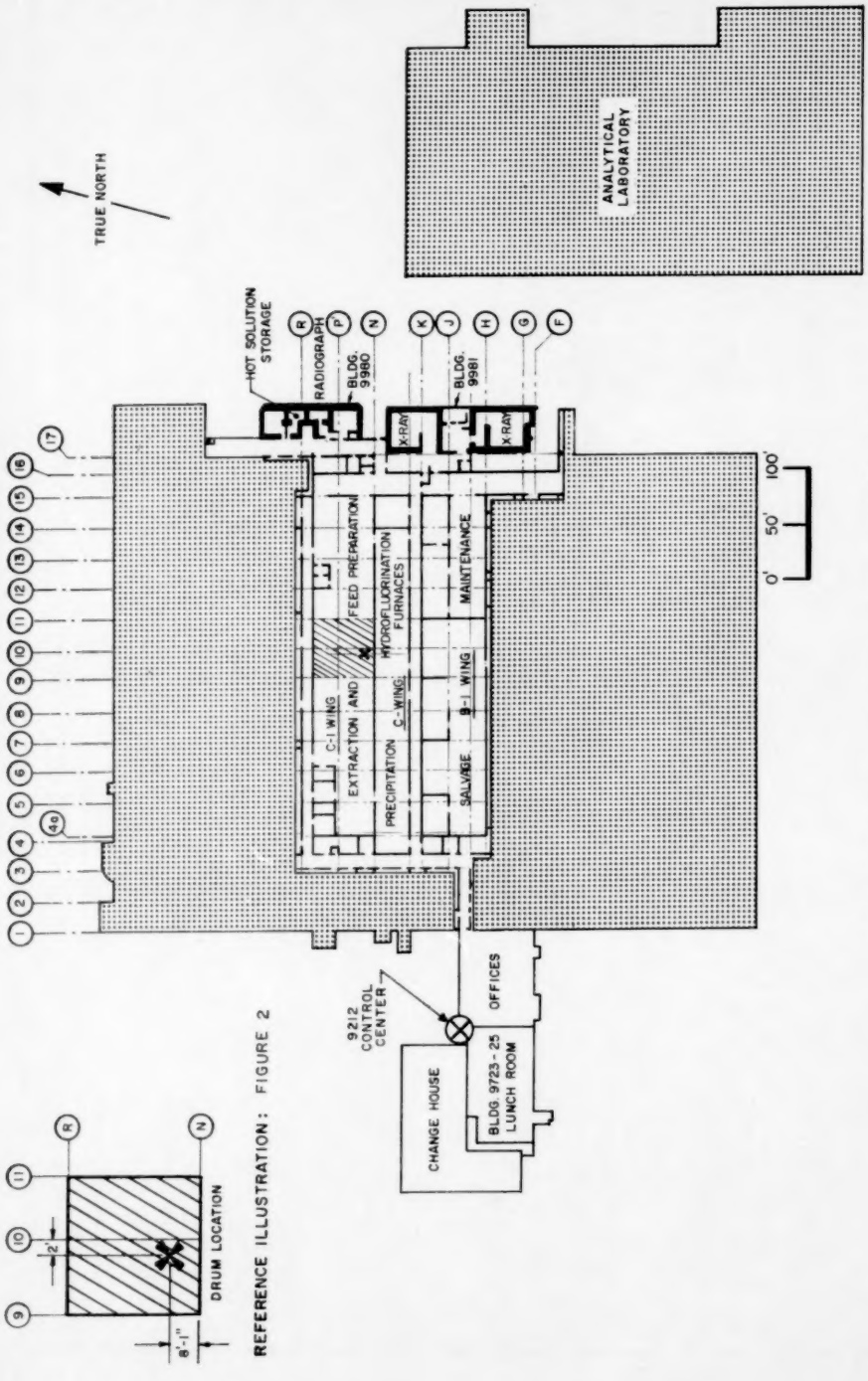


FIG. 2. Location of nuclear excursion, Building 9212.

sounded the alarm in Building 9212, implementing the building evacuation plan. Immediately, process supervisors, equipped with emergency radiation survey meters detected, and confirmed by multiple readings, radiation intensities of more than 100 mr/hr at the building control center, a distance shown later to be about 350 ft from the location of the accident. The Plant Emergency Director, who was present at the time of the alarm, acted to put the plant emergency plan into effect, thus evacuation of personnel from primary assembly points to secondary control centers proceeded with little further delay (see Figs. 1 and 2 for locations).

The local emergency squad surveyed the west and north ends of the building, observing readings of from 50 to 100 mr/hr. During this same period, radiation was detected by laboratory supervisors at the north end of the analytical laboratory, approximately 400 ft east of the accident scene, at first fluctuating in intensity up to ≈1000 mr/hr and shortly thereafter up to 500 mr/hr. It is perhaps significant to observe that the building is oriented in such a manner that the corridors and operating areas extend in an east–west direction. It is reasonable to assume that less attenuation occurs to the east and west owing to lack of walls and absence of bulky operating equipment. The area is cross-walled and heavily equipped to the north and south.

The radiation detected up to this point made it clear that the incident had occurred within Building 9212 with the precise location yet to be determined. Continuing surveys along the outside of the perimeter fence recorded readings of up to 50 mr/hr for a period of about 20 min, after which time the levels dropped to 5–10 mr/hr.

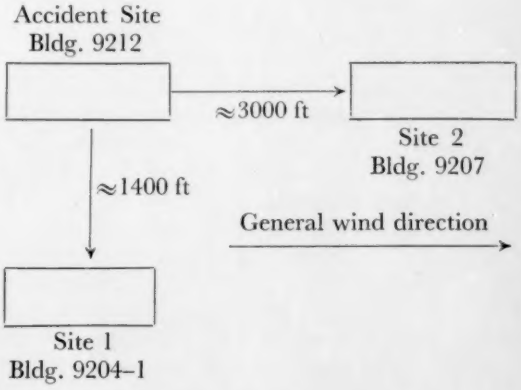
Survey of environs and detection of released activity

Radiation surveys were made of the plant area to obtain an overall evaluation of conditions. Within an hour after the incident, it was determined that there was no direct radiation or significant contamination outside the perimeter fencing, which forms the security area in which Building 9212 is located. This area, approximately 800 ft × 1000 ft, was subsequently marked off as the initial delimitation area.

Efforts were made to survey the environs and detect the release, or subsequent fallout of fission product activity. High-volume air samplers were set up outdoors at five locations ranging from ≈700 ft to ≈3000 ft downwind from the accident. Sampling was not begun until about 50 min after the incident and 20 min after the ventilation supply and exhaust fans for the area involved had been cut off. The samples, obtained by drawing air through Whatman no. 41 filters, were counted for α- and β-γ activity.

These samples indicated a maximum concentration of 2.5×10^{-11} μc/cm^3 β-γ activity as of the time of collection. This is well below the 10^{-9} μc/cm^3 permissible level of air-borne activity suggested by the National Committee on Radiation Protection.[1] As would be expected, no significant air-borne α-contamination was detected.

Some indication of air-borne contamination released to the atmosphere may be obtained from inspection of two continuously recording β-γ air monitors which were located as shown in the sketch below. These monitors are Geiger–Muller tube instruments measuring and recording the β-γ emission from particulates collected from an air stream on a fixed filter paper. The tubes are surrounded by the filter and are shielded by 1½ in. of lead.



Both instruments detected the initial direct gamma radiation from the actual excursion and both showed subsequent increases in the level of atmospheric beta–gamma contamination. The

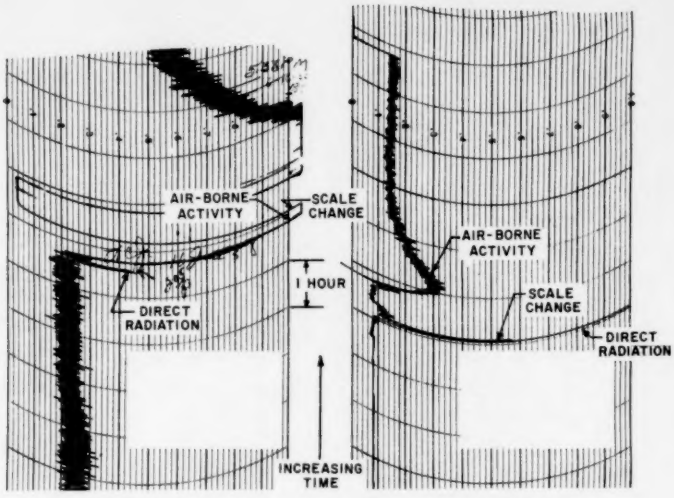


FIG. 3. Monitor chart, Building 9207 (site 2).

FIG. 4. Monitor chart, Building 9204–1 (site 1).

following observations are made from inspection of the charts from these instruments (see Figs. 3 and 4).

(1) The level of initial direct radiation recorded at site 1 was higher than that observed at site 2 because of the distances involved.

(2) The air-borne contamination, however, reached site 2 much sooner (10 min vs. ≈45 min) and in higher concentrations (considerably more than a factor of 3), since it was directly downwind.

(3) Because of the short half-lives of many of the fission products (<2 hr) and the relatively short length of exposure of any persons to the contaminated atmosphere, the concentrations detected constituted no acute hazard.

(4) Considering the favorable conditions of wind velocity and direction existing at the time and the levels of concentration detected, it can be stated, with a high degree of confidence, that no significant concentrations of these activities reached any nearby populated areas.

Surveys were made of the parking lots along the north side of the plant site, these lots being located generally east and west of the accident area. Spot checks were made on the ground, paved areas and automobiles. No evidence of β-γ contamination was detected and the automobiles were consequently released.

Survey of accident site and rehabilitation of area

Within $1\frac{1}{2}$ hr after the excursion, teams of health physicists began approximating the site of the incident by a series of perimeter radiation surveys. The radiation and contamination levels encountered were quite low, ranging from 0.2 mr/hr at the boundary of the primary delimitation arc to 10 mr/hr at distances of approximately 200 ft from the drum. Although some spillage of materials had occurred in a few areas due to unattended operating equipment, no serious contamination levels were encountered. Boundary limits were adjusted to permit employes to re-enter all areas except those within the secondary delimitation boundary (≈350 ft × 400 ft) shown in Fig. 1.

Some 3 hr after the incident, an emergency team undertook a cursory survey of the salvage area in which the reaction had occurred. The radiation dose rate itself at the southwest door of this area, a distance of approximately 100 ft from the drum, was 60 mr/hr. When the men emerged, after being in this location for about 10 min, the cannisters of their gas masks read from 10 to 15 mr/hr, indicating that significant

concentrations of air-borne contamination still existed in the area. Radiation measurements, direct and by air-sampling, were continued in the zone proximate to the accident area. Within a few hours, it was possible to relax the restriction boundaries to that area indicated as the tertiary delimitation area in Fig. 1.

Control stations, manned by health physicists and stocked with the necessary items of protective equipment, were set up in the hallways to prevent unauthorized entry into this restricted area. Authorized persons were permitted to enter the controlled zone only in teams of two or more. Each team carried at least two radiation survey instruments. Required protective equipment included coveralls, shoe covers, stocking caps, rubber gloves, and either an MSA "All Service" mask with an ultra-filter or a U.S. Army assault mask M-9 with an M-11 canister. Each person wore direct-reading pocket dosimeters and a film badge and each was surveyed for personal contamination upon return from the controlled zone.

In order to prevent the spread of air-borne contamination from the controlled zone to other parts of the building, a small fan, which exhausted through a CWS filter, was turned on, thus maintaining the area under negative pressure.

A cadmium scroll was inserted into the drum by manipulation of a 10 ft pipe, which was adapted for the purpose. This action provided positive assurance of nuclear safety and permitted a more detailed survey of the area. Such a survey was undertaken with the following results being recorded about 9 hr after the excursion:

Approximate position	Reading* (r/hr)
20 ft east of drum	1.1
12 ft east of drum	4.7
8 ft east of drum	9.8
12 ft west of drum	3.6
8 ft west of drum	8.0
12 ft north of drum	3.6
8 ft north of drum	9.8
2 ft north of drum	81.0

"Wipes" or "smears" of the floor near the drum indicated that surface contamination had been confined to the immediate vicinity of the drum. The highest activity detected on these smears was 250 mrad/hr, direct reading from the smear itself, and 16,000 d/m per 100cm^2 α-contamination. Normal α-contamination usually ranges from 500–1000 d/m per 100cm^2. The fact that the active solution was confined to the drum itself was fortunate since widespread contamination would undoubtedly have seriously hampered reoccupation and rehabilitation of the area.

The drum of poisoned solution was allowed to sit intact until the morning of the second day following the accident. During this time, additional activity measurements of the drum were made. Some indication of the decay of radioactivity is shown below:

Date	Time	Reading (r/hr)	Position
June 16	10.32 p.m.	81	at 24 in., middle of drum
June 17	10.30 a.m.	100	at 3 in., middle of drum
June 18	10.00 a.m.	48	at 3 in., middle of drum

After the immediate nuclear hazard had been disposed of by the addition of cadmium, the problem of removing the highly radioactive solution from the operating area was approached. It was decided to install "safe" tankage in an available radiographic cell some 200 ft east of the drum location and to vacuum transfer the solution via stainless steel tubing into this system, where it would be allowed to decay prior to reprocessing. Monitoring services were provided during the preparation for transfer of the solution to the shielded safe storage vessel.

* All readings were taken with an ionization chamber instrument (cutie pie) approximately 3 ft above the floor with the exception of the last reading. The reading 2 ft from the drum was made at a height above the floor of approximately one-half the height of the drum.

Dosimeters, worn by persons installing the vacuum transfer line indicated that no persons received any appreciable γ-dose (i.e. less than 300 mr) while making these preparations.

On June 18, vacuum transfer of the solution from the drum to the safe containers was begun. Radiation intensities, a few inches from the line, varied from 50 mr/hr to 80 mr/hr during transfer of the solution. During flushing of the drum and line with water, the radiation intensity dropped from 38 mr/hr during the first 5 gal flushing to 5 mr/hr during the second 5 gal flushing. Following this action, the following conditions were noted:

Empty line at exterior surface: 1 mr/hr
Top of empty drum; exterior surface: 5 r/hr
Near bottom of drum, exterior surface: 30 r/hr (due to sludge in drum)

After the active solution had been successfully transferred to shielded safe storage and the drum removed from the area, the boundaries of the control area were moved in to encompass only the operating wing itself ($\approx$50 ft $\times$ 400 ft). Contamination surveys and air samples were continued. By early afternoon of 18 June, approximately 2 days post-accident, it was determined that the exhaust ventilation fans could be turned on without significantly contaminating the surrounding areas. Subsequent air samples were well within permissible levels; personnel were then allowed to enter the accident area without respiratory protection.

Decontamination of the area proceeded as the area was released by the investigating authorities. Routine monitoring and smear surveys were made to help direct and evaluate the decontamination efforts. On 23 June, 1 week after the accident, all facilities were returned to normal operations.

SURVEY OF PERSONNEL FOR RADIATION EXPOSURES

Since 1955, strips of indium foil (approximately 1 g each) have been included in the security badges of all employees at Y-12. The purpose of these foils is to provide a quick positive means for segregating employees who receive a significant radiation dose in the course of a nuclear reaction. This determination is accomplished by the measurement of β- and γ-radiations from the radioactive In^{116} isotope which is produced by neutron irradiation of the stable In^{116} isotope in the foil.

Following assembly of personnel at the two secondary control centers, checking for neutron activation of the indium foil in their badges and for evidence of personal contamination was undertaken. Those persons whose badges gave evidence of possible high neutron doses were directed to the dispensary for further tests and medical attention. As a result of this process, twelve persons out of approximately 1200 surveyed were sent to the dispensary within 2 hr after the incident. The maximum reading observed during these checks was 60 mr/hr. In the five most significant cases, badge readings exceeded 20 mr/hr 2 hr after exposure.

In order to insure that exposure cases had not been overlooked, badges were collected at the gates when personnel were released and each badge was subsequently processed. In the course of these and the prior badge checks, about 4500 activity readings were made.

Those persons who were sent to the dispensary were checked for personal contamination, interviewed briefly, and their badges rechecked. Individuals showing evidence of β and γ body contamination were scrubbed at the dispensary decontamination facility with soap and water and mild acids.

No attempt will be made to further describe the details of the evaluation of personnel exposures since this information will be treated more fully in other parts of this series. Suffice it to say that the use of indium foil in the security badges made possible the early identification of employees who had been in the immediate vicinity of the reaction and facilitated their segregation from unexposed employees. A possible unmanageable flood of employees to the dispensary was thereby forestalled.

SUMMARY

The Y-12 critical incident was unique and began suddenly. Of particular note is the fact that the radioactive solution which comprised the reactor fuel was well contained within the reactor vessel, thus no extensive decontamination of persons or facilities was required. Further,

the distribution of fission products to the environs was limited so that no serious complications were encountered in the plant area. The seriousness of the incident was lessened by the fact that no physical violence was involved in the reaction. Rehabilitation of the area and resumption of activities were possible in a minimum amount of time. Identification and segregation of all seriously-exposed personnel was accomplished with reasonable dispatch. In this respect, the use of indium foil as an identification medium was proven to be successful.

It should not be inferred that the conditions encountered in this incident are characteristic of the consequences to be expected as a result of future nuclear accidents. Rather, in view of a number of unique, fortunate circumstances which reduced the Y-12 problem significantly, it is reasonable to assume that consequences in other cases could be much more severe.

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Cs^{137} BIOSPHERIC CONTAMINATION FROM NUCLEAR WEAPONS TESTS*

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Abstract—Weapons tests to date (end of 1957) have produced about 6 and 11 Mc of Sr^{90} and Cs^{137}, respectively, of which about 1.9 and 3.4 Mc have been deposited as long-range fallout. Average surface deposition levels of Sr^{90} and Cs^{137} in the north temperate population belt may be about 20 and 36 mc/mile^2, respectively, and the stratospheric reservoir may contain an additional 2.3 Mc of Sr^{90} and 4.2 Mc of Cs^{137}.

Cesium-137 contamination of the atmosphere and of drinking water, assuming they are in equilibrium with the rate of stratospheric fallout, does not appear to be a serious potential hazard to man's health even though weapons tests continue indefinitely at the past rate. The ingestion of contaminated foods is the principal source of Cs^{137} in the population. At present it is not possible to determine whether cesium is entering the biosphere and man via direct contamination of vegetation or via soil integration and plant uptake. If its mode of entry is through the soil, the potential radiation hazard from continued tests may be a factor of about 4 greater than if it is entering by direct contamination of vegetation.

Because of metabolic similarity of cesium and potassium, cesium levels in the biosphere are conveniently expressed as Cs^{137}/g K. The average 1957 Cs^{137} level in United States milk was about 32 pc†/g K, and the level in the rest of the diet was about 20 pc/g K. The average 1956, 1957 and 1958 (through July) levels in the United States population were about 41, 44 and 54 pc/g K, respectively.

Correlation of Cs^{137} levels in the population with levels in milk suggests a dietary discrimination factor of about 2 in favor of cesium over potassium in going from the diet to the body. Correlation of 1957 Cs^{137} levels in United States milk (expressed as pc/g K per in. of rainfall) with geographic meteorological conditions suggests that the manner in which tropospheric air masses approach or traverse the latitudes of high stratospheric fallout may be a factor in Cs^{137} and Sr^{90} surface deposition levels. The increase in Cs^{137} levels in milk and in the United States population from 1956 to 1957 was not proportional to the increase in integrated fallout, suggesting direct contamination of vegetation as its route of entry into the biosphere. However, measured levels presently in the population are compatible with predictions based on ecological considerations.

The average 30 year internal plus external Cs^{137} radiation dose in equilibrium with a continuing test rate of 10 MT of fission per year is estimated as about 300 mr, or about 10 per cent of natural background, and the average total 70 year bone marrow dose is estimated as about 500 mr (assuming Cs^{137} enters the biosphere via the soil). Assuming 30–50 r as the genetic doubling dose and a non-threshold response to radiation, indefinite testing at the rate of 10 MT of fission per year may produce 0.5 to 1 per cent increase in the incidence of mutations as a result of Cs^{137} biospheric contamination. If weapons tests are stopped, the increase in incidence of mutations from Cs^{137} would be about one-tenth of the above estimate. A comparison of the mean bone marrow dose from Sr^{90} and Cs^{137} suggests that

* Work performed under the auspices of the U.S. Atomic Energy Commission.
† pc is used throughout to indicate micro microcurie ($\mu\mu$c).

Cs^{137} may pose a potential leukemogenic hazard comparable to Sr^{90}, if both are entering the biosphere via soil integration and plant uptake. If Cs^{137} is entering the biosphere via direct contamination of vegetation, its potential leukemogenic hazard is about one-fifth that of Sr^{90}. Present knowledge is not adequate to establish the nature of the dose–response relationship for somatic effects of radiation. Therefore, it is not possible at present to estimate the absolute leukemogenic hazard of either Sr^{90} or Cs^{137} fallout.

INTRODUCTION

Testing of nuclear weapons releases radioactive debris into the atmosphere, and its deposition over the earth's surface is termed "fallout". The net result of fallout is a small increase in the external and internal radiation background to which all life is exposed. Increased external exposure may augment the incidence of genetic changes. Assimilation of radionuclides may result in either somatic or genetic damage, depending upon their site of body deposition. The problem of the potential hazard of fallout then becomes one of ascertaining the magnitude and significance of this increase in background radiation dose with regard to its potential risk to man's health and well-being.

Fallout is divided into three categories (local, intermediate or tropospheric, and stratospheric or world-wide) on the basis of time and place of deposition.

Local fallout is deposited within the first 24 hr and confined to the immediate environs of the detonation. Tropospheric fallout occurs in from 30 to 60 days essentially in a west-to-east direction in the general latitude of the detonation. Stratospheric fallout can result only from large weapons and may require years to be deposited ($T_{1/2} \sim 4$–5 years). Its distribution is essentially world-wide, although there are strong indications of preferential deposition in the temperate latitudes.

This report is restricted mostly to the potential internal hazard of distant (tropospheric plus stratospheric) fallout from nuclear tests, with emphasis on Cs^{137}. Under these conditions, only the long-lived isotopes, Sr^{90} and Cs^{137}, are important. Strontium-90 is treated only superficially since it has been discussed in previous publications.[1,2]

Cesium-137 appears relatively less important than Sr^{90} in terms of integrated radiation dose. However, Sr^{90} is primarily a somatic hazard, whereas Cs^{137} can produce both somatic and genetic effects. Until our knowledge of the potential hazards of chronic, low-level radiation exposure is more quantitative, Cs^{137} should not be ignored.

PRODUCTION AND DISTRIBUTION OF Cs^{137} AND Sr^{90}

An estimation of the production of Cs^{137} and Sr^{90} from past tests would be helpful in assessing the potential hazard of present biospheric contamination and in extrapolating to future levels in the event of continued testing or nuclear war.

Statements during the Subcommittee Hearings before the Joint Committee on Atomic Energy, Congress of the United States[3] assumed a constant test rate of 10 MT of fission per year, beginning in the spring of 1952. This leads to a total testing by all nations of about 60 MT of fission yield by the end of 1957. Libby[4] has estimated the periodic increments of fission debris added to the stratosphere (Fig. 1) and has listed the contribution of the various countries to the total tropospheric and stratospheric reservoirs (Table 1). These data show that total stratospheric contamination to the end of 1957 was about 36 MT equivalents of fission yield, about 60 per cent of which was contributed by the United States Castle test series in the spring of 1954. Total tropospheric contamination by all countries was given as 5.6 MT equivalents, and the total tropospheric plus stratospheric as about 42 MT equivalents.

If a total fission yield of 60 MT to the end of 1957 and the above data are assumed to be reasonably realistic, then the local, tropospheric and stratospheric injection rates over the past 6 years have averaged 30, 10 and 60 per cent, respectively. For individual detonations, of course, distribution of debris is determined by weapon yield and by conditions of firing. The megaton devices detonated at the United States

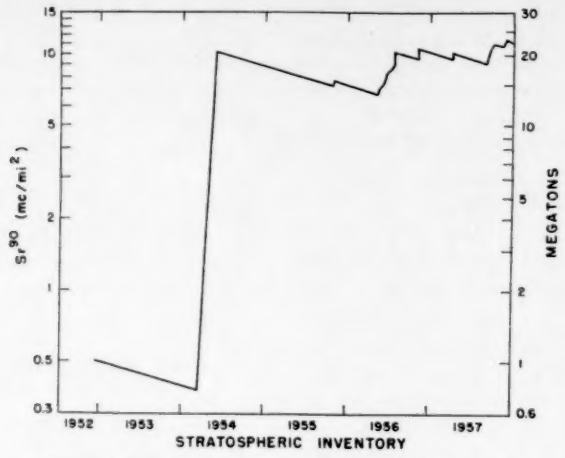

FIG. 1. Stratospheric inventory and rate of fallout of fission debris.[5]

Pacific Test Site have been fired under conditions that have maximized local fallout and minimized atmospheric contamination.

MACHTA and LIST[5] have estimated the rate of stratospheric fallout at about 20 per cent per year, as compared to LIBBY's estimate of 10 per cent. MACHTA's application of a 20 per cent fallout rate to LIBBY's data on stratospheric injection (Fig. 1) leads to an estimate of 13 MT equivalents of fission debris deposited from the stratosphere by the end of 1957 and a remaining stratospheric inventory of 23 MT equivalents. From these data total stratospheric plus tropospheric deposition would, therefore, appear to be about 18.6 MT equivalents of fission debris at the end of 1957.

Table 1. *Sources of radioactive fallout to December* 1957 (from MACHTA and LIST,[5] quoting LIBBY)

Country	Site of production (latitude)	Equivalents of fission yield (MT)
Contribution to troposphere		
U.S.S.R.	50°N	1.7
U.S. (Nevada)	37°N	1.0
U.S. (Pacific)	11°N	1.3
U.K. (Pacific)	3°N	1.5
U.K. (Australia)	35°S	0.1
		Total 5.6
Contribution to stratosphere		
U.S.S.R.		11.2
U.S.		24.8
		Total 36.0
Total tropospheric and stratospheric contribution		41.6

As 1 MT of fission energy release produces approximately 0.1 megacurie (Mc) of Sr^{90}, the total world production (by the end of 1957) is about 6 Mc. Radiochemical analyses of fission debris suggest that about 0.18 Mc of Cs^{137} is produced per megaton of fission energy release. From these data and the previous discussion, the production and distribution of Cs^{137} and Sr^{90} may be estimated (Table 2) assuming no fractionation has occurred. These data indicate a world average surface deposition level (December 1957) of about 9 and 17 mc/mile^2 for Sr^{90} and Cs^{137}, respectively. The stratospheric reservoir contains an additional 12 mc/mile^2 of Sr^{90} and 21 mc/mile^2 of Cs^{137}. The actual content of the stratosphere, of course, is highly uncertain, and there is some reason to believe it may contain more nearly 12 MT equivalents than 24.[2]

The assumption of no fractionation of the various components of fission debris between the time of production and deposition is not supported strictly by experimental observation. STEWART *et al.*[6] reported an average Cs^{137}/Sr^{90} ratio of 1.5 in rainwater samples collected in

Table 2. Production and distribution (assuming no fractionation) of Sr^{90} and Cs^{137} from nuclear detonations (December 1957)

Distribution	Sr^{90} (Mc)	Cs^{137} (Mc)
Total production	6	10.8
Local deposition	1.8	3.2
Tropospheric deposition	0.6	1.1
Stratospheric deposition	1.3	2.3
Total tropospheric plus stratospheric deposition	1.9*	3.4
Stratospheric reservoir	2.3*	4.2

* EISENBUD estimated 1.9 and 1.8 Mc Sr^{90} for long-range fallout and the stratospheric reservoir, respectively. His estimates were based on data from the world-wide gummed film network (Notes on Deposition of Sr^{90} in Mid-1957, in manuscript).

various parts of the world during 1955–1957. Although the ratios varied greatly, the observed average compares reasonably with their ratio of production (1.8), and as a general approximation the assumption of no fractionation may be acceptable.

Essentially no data are available on Cs^{137} integrated soil deposition levels. Recently, however, MACHTA[5] estimated the latitudinal deposition level of Sr^{90} (lower curve, Fig. 2) as of December 1957 by adjusting the 1956 soil analysis data on the basis of New York fallout measurements made from steel pot collections. If it is assumed that the Cs^{137}/Sr^{90} deposition ratio is the same as their ratio of production, then the latitudinal distribution of integrated Cs^{137} deposition is represented by the upper curve of Fig. 2.

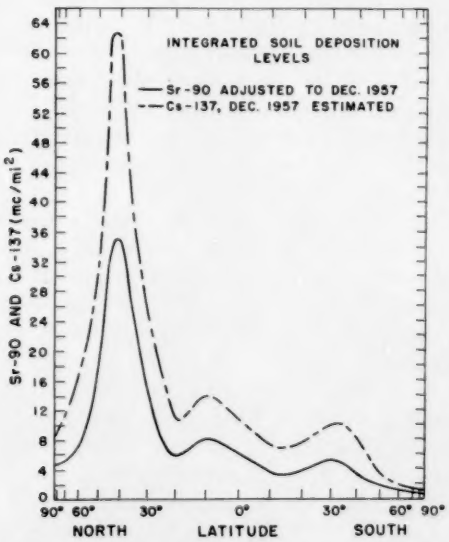

FIG. 2. Latitudinal distribution of Sr^{90} and Cs^{137} fallout.[5]

These curves are very general and obviously were not intended to consider fluctuations as a result of local meteorological factors. They suggest that the expected general average integrated Sr^{90} and Cs^{137} surface deposition levels (as of December 1957) in the north temperate population belt (~10°N–60°N latitude) may be about 20 and 36 mc/mile^2, respectively. Respective general maximum values near 40°N–45°N latitude may run as high as 35 and 60 mc/mile^2. The high values observed in the temperate latitudes appear to be explained more satisfactorily by the preferential stratospheric fallout model proposed by MACHTA[5] and STEWART *et al.*[6] than by the uniform stratospheric fallout theory proposed by LIBBY.[7]

INCORPORATION OF Cs^{137} INTO THE BIOSPHERE

When weapons debris is released to the earth's environment, it may enter the biosphere through direct intake and through intake via the soil–plant or ecological cycle. Eventually it finds its way into man through inhalation of contaminated atmosphere and ingestion of contaminated food and water.

Regardless of mode of entry, the various radionuclides enter the biosphere and man according to their individual chemical, biological and physical properties. Cesium is chemically and metabolically similar to potassium, an essential body constituent. It seems reasonable, therefore, to consider its incorporation into the biosphere in terms of Cs^{137}/K ratios. Natural potassium is also radioactive, and measurement of the natural isotope, K^{40}, provides a direct means of establishing Cs^{137}/K ratios. Highly important also is the fact that

radioactivity of natural body potassium delivers a total body radiation dose of about 20 mr/year, or about 20 per cent of man's natural radiation background.

Cs^{137} *contamination of the atmosphere*

MARTELL[8] measured the mean Sr^{90} content of ground level air at Washington, D.C., during 1952–1955 and reported a value of 3×10^{-15} $\mu c/cm^3$, which is in reasonable agreement with the mean value of 4×10^{-16} $\mu c/cm^3$ calculated by STEWART *et al.*[9] for the same period in England. The ratio of Cs^{137}/Sr^{90} in rainwater (and, therefore, in air) was measured as 1.5.[6] The mean concentration of Cs^{137} in air during the same period, therefore, would have been about 5×10^{-15} $\mu c/cm^3$. The maximum permissible concentration of Cs^{137} in air, based on ICRP standards[10] for continuous exposure of the general population and a biological half-time of 140 days, is about 3×10^{-9} $\mu c/cm^3$, about 500,000 times the observed value. Since the tropospheric fallout time is 30–60 days, the mean air concentration during 1952–1955 probably approximates equilibrium conditions with the past 5 year rate of stratospheric fallout. In this case, continued weapons testing at the average rate of the past 5 years should not increase the mean air concentration and the inhalation risk significantly. If air containing 5×10^{-15} $\mu c/cm^3$ of Cs^{137} is breathed indefinitely, the equilibrium total body burden of the standard man would be only about 0.07 pc/g K (total body burden 10 pc) compared to an observed average burden in the United States population of 44 pc/g K in 1957. Only about one-six-hundredth of the present Cs^{137} in people could be a result of direct inhalation. As suggested by BRYANT *et al.*,[11] inhalation of nuclear debris (and Cs^{137}, specifically) does not appear to be a major factor in the potential hazards of world-wide fallout.

Cs^{137} *contamination of water*

STEWART *et al.*[6] reported Sr^{90} concentrations in rainwater collected in the north temperate latitudes during 1955-1957 averaged about 3.3 pc/l. and had an average Cs^{137}/Sr^{90} ratio of 1.5. The mean Cs^{137} content, therefore, was about 5 pc/l. or 5×10^{-9} $\mu c/cm^3$. This value is about one-three-thousandth of the maximum permissible concentration for continuous consumption based on ICRP standards[10] for the general population and an effective biological half-time of 140 days.

Since the radioactive content of rainwater is also dependent on the content of tropospheric air, which is dependent on stratospheric fallout rate, the value of 5×10^{-9} $\mu c/cm^3$ may be considered an equilibrium value with the past weapons test rate. In this case, continued testing at the past 5 year rate should not increase the potential hazard from Cs^{137} contaminated water. At equilibrium with the average Cs^{137} content of raw rainwater, the average population body burden would be about 7 pc/g body K or about one-sixth of the observed 1957 average level. However, most drinking water is derived from surface water from which Cs^{137} (and Sr^{90}) have been removed by the ion exchange system of soils. Therefore, the above calculation gives an extreme upper limit. In any event, the foregoing discussion shows that the principal route of entry of Cs^{137} into man must be through the ingestion of contaminated foods.

Cs^{137} *contamination of foods*

When Cs^{137} is deposited on the earth's surface and incorporated into the soil, it is taken into plants through the root system according to its specific soil–plant relationships. That which settles directly on vegetation may remain as surface contamination or may enter the plant through foliate absorption. When contaminated plants are eaten by animals and man, the Cs^{137} is absorbed and deposited in the body according to its own specific metabolic properties and in relation to body potassium. Since consumption of contaminated foods is the principal route of entry of Cs^{137} into man, measurements of the Cs^{137}/K ratios in food products are important.

In the spring of 1956, the Los Alamos Scientific Laboratory began an extensive survey of the Cs^{137}/K ratio of the United States milk supply. Measurements are made by γ-assay in a large volume 4π liquid scintillation counter with the capability of simultaneously measuring the 0.661-MeV γ-ray of Cs^{137} and the 1.46-MeV γ-ray of K^{40}.

FIG. 3. Geographic distribution of Cs^{137} levels in United States milk during 1957, expressed as pc/g K (numbers in parentheses represent total samples analyzed).

The average Cs^{137} content of dried or fresh milk samples collected from eleven stations in various sections of the United States was 24 pc/g K[12] during 1956. The number of samples and locations, however, were too small to constitute a general average of the nation's milk supply. During 1957 and again in 1958, the number of sampling areas and frequency of sampling were increased greatly. During 1957, dried milk was routinely sampled from thirty-three locations within the United States, and a few spot-checks were made on foreign milk.[13] A total of 887 measurements were made. The 1957 levels of Cs^{137} in milk from the various parts of the United States are shown in Fig. 3. Changes in Cs^{137} in milk from a given locality throughout the year showed indications of seasonal variation on which was superimposed transient peaks due to tropospheric contamination during periods of weapons testing. The average Cs^{137} content of United States milk samples was 32 pc/g K and varied from 59 pc/g K in Washington state to 12 pc/g K in lower California and Arizona (the areas of lowest rainfall). Single samples from England and Japan (produced late in 1956) both had a Cs^{137} content of 25 pc/g K. Five samples from Argentina (produced in November and December 1956) had an average Cs^{137} content of 9 pc/g K with a range of from 4 to 12. The average was a factor of 4 lower than comparable samples from the northern United States. Twelve monthly samples from Australia during 1957 did not indicate such a large hemispheric effect, as they showed an average Cs^{137} content of 18 pc/g K compared to from 20 to 40 for areas of similar annual rainfall in the United States.

Fig. 4 shows a plot of the average Cs^{137} in milk against the total precipitation for 1957. The milk average is computed for those periods in which tropospheric contamination was absent as indicated by the absence of Ba^{140}. The

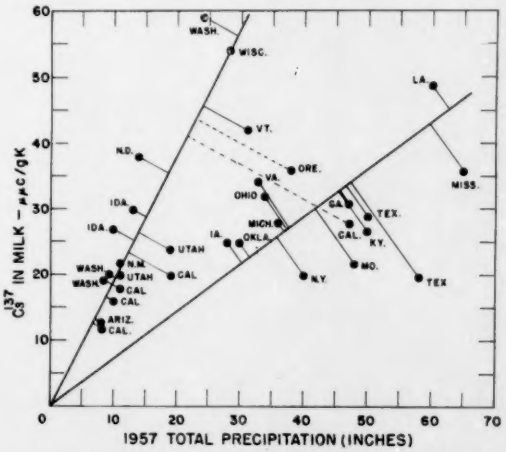

FIG. 4. Correlation of 1957 Cs^{137} levels in milk with respect to total rainfall.

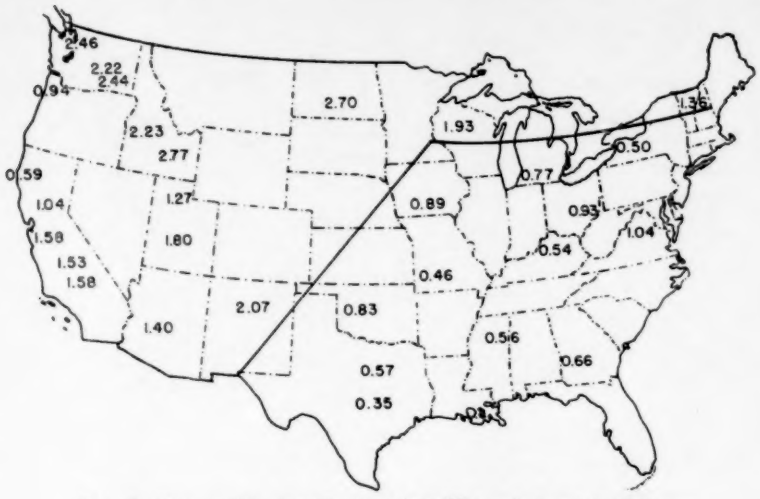

FIG. 5. Geographic distribution of Cs^{137} levels in United States milk during 1957 (pc Cs^{137}/g K per in. of rainfall).

months of June, July, September and October were principally affected. The data appear to divide into two groups, each group showing a fair correlation between cesium level and rainfall, but with pronounced difference in slope. One group includes the far western and northernmost states; the other the midwest, south and east.

Fig. 5 shows the same data expressed as pc Cs^{137}/g K per in. of precipitation superimposed on a map of the United States. A line from El Paso, Texas, to Minneapolis, Minnesota, and then due east divides the country into two regions, one having a higher value of Cs^{137}/g K per in. of rainfall and the other having a lower value. (Two outstanding anomalies in the former region are Portland, Oregon, and Fernbridge, California.*) The pattern suggests the possibility that the two regions may be a reflection of the general derivation of their precipitation, the Pacific Ocean in one case, and the Gulf of Mexico and the Atlantic Ocean in the other. The source of the water is, of course, irrelevant but the manner in which the tropospheric air masses principally responsible for precipitation approach or traverse the latitudes of high stratospheric leakage may be a factor. A similar correlation between Sr^{90} fallout (mc/mile^2) and rainfall has been pointed out by MACHTA[14] and COLLINS and HALLDEN.[15]

Through July of 1958, 840 milk samples have been analyzed. For United States stations, the average Cs^{137} content of 1958 milk to date is 35 pc/g K, with a spread of from 5 to 90. During the spring of 1958, milk samples were marked by indications of unusually heavy tropospheric contamination, due presumably to Russian spring test operations. Ignoring short-term tropospheric peaks, the average Cs^{137} content of milk for the first half of 1958 was 15 pc/g K in the southwest and 30 pc/g K in the northwest, except for the high rainfall coastal area which averaged 45. The midwest and east were quite uniform at about 35; the southeast averaged 50 pc/g K. The highest values were found in northern Wisconsin, Minnesota and North Dakota, where the

* The anomalous results in these regions may be explained on the basis of orographic precipitation. The prevailing onshore winds are lifted by the coastal ranges, causing clouds and precipitation. Such clouds do not normally extend to as great heights as those associated with the cyclonic storms dominating the precipitation patterns elsewhere in the western states. This should result in less scavanging per inch of rainfall of radioactivity from the higher concentrations found aloft. (We are indebted to Dr. R. J. LIST of the U.S. Weather Bureau for this interpretation.)

Table 3. Cs^{13} Content of miscellaneous foodstuffs[12]

Sample and date	Cs^{137} (pc/g K)
Meats—spring 1956	
Beef rounds	42
Lambs, dressed carcass	65
Pork, fresh hams	24
Pork, loins	15
Meats—winter 1956-1957	
Beef sirloins	12
Lamb, dressed carcass	13
Pork, loins	8
Pork, loins	6
Flour—spring 1956	
Wheat, Colorado	7
Bleached, enriched	26
Bleached, enriched	22
Whole wheat, graham	9
Potatoes—spring 1956	
Colorado	< 5
Idaho	< 5
Vegetables—spring 1956	
Lettuce	< 6
Cabbage	10
Carrots	< 2
Fruits—spring 1956	
Tomatoes	3
Oranges	30
Grapefruit	20
Watermelon	< 2
Coffee—spring 1956	< 4

averages reached 90 pc/g K. A population-weighted average for the entire United States would be about 35 pc/g K.

Little information has been reported on the Cs^{137} of foods other than milk. ANDERSON *et al.*[12] reported a few results on foods collected during the spring of 1956 and the winter of 1956–1957. These results are shown in Table 3.

Some indication of the Cs^{137} content of the general 1957 United States diet, excluding milk, may be inferred from a plot of 1957 milk values against the average Cs^{137} content of people from the same areas for the same period (Fig. 6). These data show a definite positive correlation between the Cs^{137} activity in milk and in people. The slope of the regression line is 0.7. If about 40 per cent of dietary potassium is derived

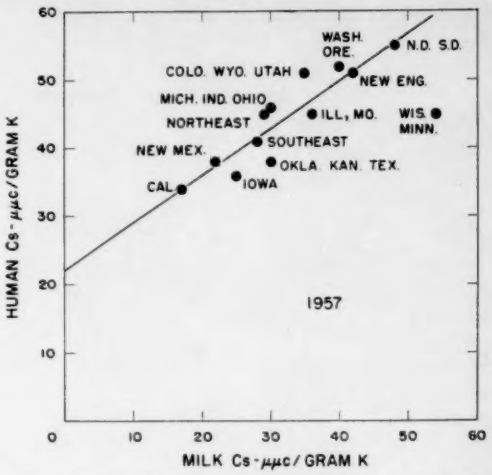

FIG. 6. Correlation of Cs^{137} levels in the United States population with levels in milk (1957).

from milk, then the discrimination factor (i.e. the ratio of $(Cs^{137}/K)_{man}$ to $(Cs^{137}/K)_{milk}$) for Cs^{137} from milk to man is 1.8. Extrapolation of the line to no cesium in milk indicates that about 23 pc of Cs^{137}/g K in people must have been derived from the rest of the diet. Assuming 60 per cent of dietary potassium from non-milk sources and a discrimination factor of 1.8, the Cs^{137}/K ratio in the rest of the diet must have been about 21 pc/g.

Another estimate of the discrimination factor can be obtained from the estimate of average dietary Cs/K given in Table 4. Since some uncertainty attaches to the average Cs/K ratios given in column 3, the resulting dietary average is only approximate. However, combining this dietary value with the observed human average of about 40 pc/g during the same period indicates a discrimination factor of about 2.

MILLER and MARINELLI[16] estimated a dietary discrimination factor of about 2 on the basis of the ratio of excretion to body burden. The dietary discrimination factor calculated from the ratio of the biological half-times of cesium and potassium (discussed later) is about 3.5.

Cs^{137} *contamination of the population*

Metabolism of Cs^{137} *in man and animals.* The absorption, retention and excretion of orally

Table 4. Estimated Cs^{137} content of average diet

Source	Dietary potassium (%)	Average Cs/K 1956-1957 (pc/g)	Contribution to dietary Cs (pc/g K)	(%)
Dairy products	38	32	12	60
Meat	15	32	5	25
Flour and cereals	6	24	1.4	7
Vegetables	18	5	0.9	4
Citrus fruits	3	25	0.7	3
Potatoes	19	0	0	0
Average Cs/K				20

administered tracer doses of Cs^{134} or Cs^{137} were studied in five animal species[17] using *in vivo* whole-body counting methods. Absorption of radiocesium by mice, rats, dogs, monkeys and man was essentially 100 per cent. The whole-body retention curves for the five species are shown in Fig. 7. The retention equations are not simple exponentials but are multiple rate functions with three principal components (only two established so far for man). The radiocesium retention equation for man over at least 650 days was

$$R_t = 16 \exp\left\{\frac{-0.693}{3} t\right\} + 84 \exp\left\{\frac{-0.693}{143} t\right\}$$

where R_t is the percentage retained, and t is the time after administration in days. From analogy with the other species, a third component (representing a small fraction of the dose) to the rate equation for man may be expected at a later time. The biological half-time of the major component of the cesium retention function is 143 days, and the corresponding half-time for potassium retention has been estimated as about 40 days.[17] The discrimination factor for Cs^{137} with respect to potassium (under conditions of chronic exposure) in going from diet to man should, therefore, be $143/40 = 3.5$, which is not consistent with the value of about 2 derived from the measurements and dietary considerations given earlier.

An explanation for this apparent anomaly is that potassium may not have a unique half-time in the body. The turnover time of potassium, like that of body water, may be governed entirely by the intake rate, the amount retained being homeostated by the minimum and maximum requirements of the body. Cesium-137, on the other hand, appears to be excreted at a constant rate independent of the potassium intake rate so long as the latter does not exceed reasonable limits.[18,19]

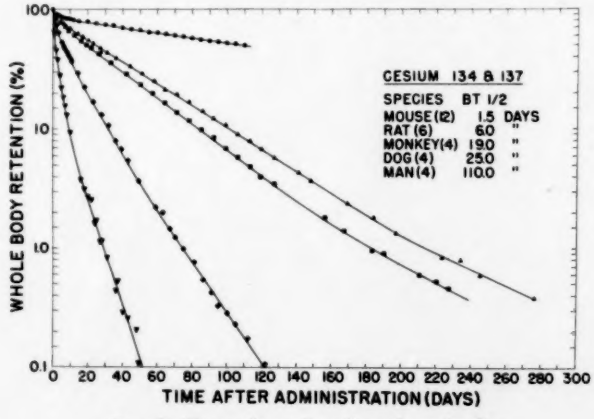

FIG. 7. Retention of radiocesium in five mammalian species.

HOOD and COMAR[20] reported retention half-times for cows (~20 days) which were considerably shorter than those observed for man. Their data indicate ruminants have an unusually high fecal excretion rate compared to single-stomach species and lactating cows lose sizeable quantities in milk (~12 per cent in 30 days). In the absence of specific data, there is no choice but to assume that cesium and potassium would act similarly in ruminants and that the discrimination factor for cesium with respect to potassium in going from diet to tissues of domestic animals would be comparable to that for other species.

Cesium, like potassium, is retained principally in muscle. Rats sacrificed 5–20 days after oral administration of Cs^{134} had 80 per cent of their total body burden in the muscle mass.[17] Apparently about 80 per cent of the body burden is confined to the muscle mass in cattle, sheep, swine[20] and man[17] also. Concentration of radiocesium in the gonads of rats 2 days after administration was about 70 per cent of that in muscle, and it rate of elimination from reproductive tissues was faster than from muscle.

Present Cs^{137} *levels in people*

The Cs^{137} level in the United States population has been studied at the Los Alamos Scientific Laboratory since the spring of 1956. The method used is *in vivo* whole-body counting in a 4π liquid scintillation counter. Discriminator settings enabled simultaneous determination of Cs^{137} and K^{40}, and consequently the determination of the total body Cs^{137}/K ratio. In 1956, 196 determinations were made on people from twenty-seven states and during 1957, 311 determinations were run on people from thirty states. During 1958 to 1 August, 1100 measurements covering thirty states have been made. Summaries of the 1956[12] and 1957[13] results have been published elsewhere.

The average Cs^{137}/K ratio in the United States population was 41 pc/g during 1956 and 44 pc/g in 1957. The average during 1958 to date is 54 pc/g. There appeared to be no striking difference between the 1956 and 1957 averages. However, there does seem to be a significant increase in the 1958 levels. The frequency distribution curves for each of the 3 years appeared normal with standard deviations of 36 per cent. The 1957 Cs^{137} data for the United States population as a function of geographic location are shown in Fig. 8. The highest values were found in the northern region and the lowest in southern regions; however, the differences were not great—in keeping with expectation because of the rather wide food distribution system in the United States. The correlation of 1957 Cs^{137} data for people with that for milk was shown earlier in Fig. 6, and a positive correlation is definitely indicated. This also may be expected since milk is unique in its localized consumption.

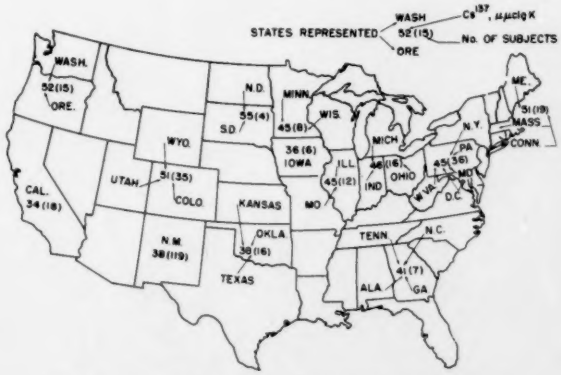

FIG. 8. Geographic distribution of Cs^{137} levels in the United States population (1957).

Table 5. Levels in people

Country	Year	Number of measurements	Cs^{137} (pc/g K)	Reference
North America				
USA	1956	196	41	(12)
USA	1957	311	44	(13)
USA (Chicago)	1957	13	35	(16)
USA (Utah)	1957–1958	42	66	(21)
USA	1958	1100	54	(22)
USA (New Mexico)	1958	500	50	(22)
Canada	1957	4	66	(22)
South America				
Colombia	1957	1	21	(16)
Ecuador	1957	1	13	(16)
Bolivia	1957	1	13	(16)
Brazil	1957	2	19	(16)
Uruguay	1957	1	11	(16)
Argentina	1957	2	8	(16)
Chile	1957	1	22	(16)
Central America and West Indies				
El Salvador	1957	1	8	(16)
Puerto Rico	1958	1	40	(22)
Cuba	1957	1	20	(16)
Europe				
England	1956	2	35	(16)
England	1956–1957	16	34	(23)
England	1957	2	50	(22)
England	1958	7	59	(22)
France	1956	1	33	(16)
France	1957	1	63	(22)
France	1958	2	55	(22)
Belgium	1956	1	27	(16)
Sweden	1956	1	32	(16)
Sweden	1957	3	52	(22)
Sweden	1958	2	62	(22)
Germany	1957	2	56	(22)
Germany	1958	2	52	(22)
Australia				
	1957	1	50	(16)
	1958	1	50	(22)
Asia				
India	1956	2	20	(16)
Japan	1956	1	25	(16)
Japan	1957	1	30	(22)
Japan	1958	1	40	(22)
Indonesia	1956	2	11	(16)
Thailand	1957	3	34	(22)
Iran	1958	1	53	(22)
Neapl	1958	1	35	(22)
Africa				
South Africa	1958	2	44	(22)

Others have reported Cs^{137} levels in people, and these results are summarized and compared in Table 5 with those collected by the Los Alamos Scientific Laboratory.[12,13,22] The data for people outside the United States are too few to give any indication of world-wide geographic distribution other than to say that there definitely seems to be a hemispheric difference.[16] The statistical and sampling uncertainties in the results on people outside the United States are large because of the small number of determinations and the method of sampling. Only people traveling in the United States were measured and family groups were occasionally included. Both of these factors may favor a non-random sampling bias. If it is assumed that sampling was random and that the frequency distribution of results was the same as that observed for the United States, the standard deviation of a single determination would be ± 36 per cent. The standard deviation of the mean for the sixteen foreign determinations during 1957 would be at least 9 per cent. The 1957 average burden of 52 pc/g K for people outside the United States, therefore, is not significantly different from the United States average of 44. A preliminary estimate of the 1958 United States average is 54 pc/g K, compared to a 1958 average of 56 for persons outside the United States.

The data suggest that the 1957 average Cs^{137} body burden in the north temperate population belt (and thus in the majority of the world's population) may be about 40 pc/g K. Cesium-137 levels in residents of Utah (essentially all from Salt Lake City) definitely seems higher than the United States average. The suggestion has been made that this is a real observation and that it is a result of local fallout from the Neveda Test Site.[21] MILLER and MARINELLI[16] have suggested also, on the basis of the Asian subjects who consume little or no dairy products, that a dietary difference is indicated.

Ecological incorporation of Cs^{137}

Cesium is chemically and metabolically similar to potassium, and its uptake via the ecological cycle and incorporation into man should be in relation to the exchangeable or available soil potassium. Exchangeable soil potassium, to a depth of 2.5 in., may vary from about 25 to 400 lb/acre. About 100 lb/acre is a reasonable average value for the agricultural soils of the United States. This is equivalent to about 3×10^7 g of exchangeable K/mile^2. Deposition and mixing to a depth of 2.5 in. of 1 mc of Cs^{137}/mile^2 gives a total concentration of about 30 pc/g of exchangeable soil potassium. However, when Cs^{137} comes in contact with soil, it is rapidly fixed. Leaching studies[24] show essentially all of the Cs^{137} remains in the top inch of soil, even after 200 in. of simulated rainfall. The extent of fixation, as with potassium, is proportional to the colloidal content of the soil.

LARSON *et al.*[25] added Cs^{137} to three different types of soils and determined the amount that could be extracted with 1 N NH_4Ac. The exchangeable Cs^{137} ranged from 13 to 33 per cent with an average of 25.

Plants discriminate heavily against Cs^{137} with respect to potassium, even when the cesium is in an exchangeable form. AUERBACH[26] reported uptake of Cs^{137} by corn grown in a lake bed once used for the disposal of reactor wastes. He found that the Cs^{137}/K ratio in the plants was about 1 per cent of the exchangeable Cs^{137}/K ratio in the soil. MENZEL[27] obtained a discrimination factor of about 0.04 between Cs^{137}/K in barley and corn and the ratio in available soil potassium, and definitely showed that plant uptake of Cs^{137} was inversely proportional to exchangeable and available soil potassium. Without considering exchangeable soil potassium, others[28,29] have studied the ratio of Cs^{137} per gram of dry plant materials to the concentration per gram of soil and obtained values of 0.006 to 0.18 (average 0.07). These data suggest that the average Cs^{137} concentration in the potassium of plants should be about 0.04 times the exchangeable Cs^{137} concentration in exchangeable soil potassium. Assuming 75 per cent of the deposited Cs^{137} is fixed in a form unavailable to plants, the discrimination factor,

$$\frac{(Cs^{137}/K)_p}{(Cs^{137}/K)_s},$$

against total cesium with respect to available soil potassium, in going from soils to plants, is about $0.01(DF_1)$. If plant uptake of Cs^{137} was

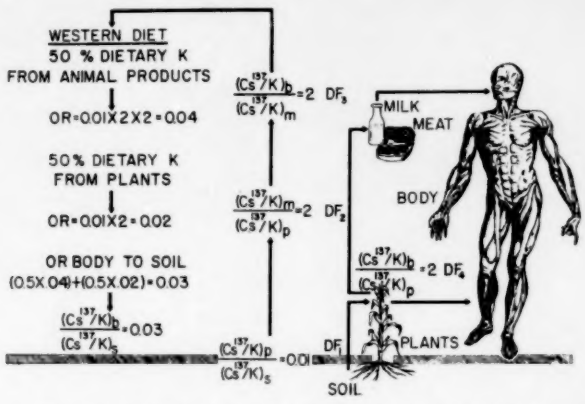

FIG. 9. Ecological discrimination against Cs^{137} with respect to potassium (United States population).

via the soil, the average 1957 Cs^{137} content of plants in the north temperate population belt would be estimated at about 10 pc/g K.

The discrimination factor in going from diet to man, derived earlier, was about 2. If it is assumed that a factor of 2 applies also to Cs^{137} in going from plants to animal products (which provides about 50 per cent of man's dietary potassium), the overall discrimination ratio in going from soil deposition to man is estimated as about 0.03 (Fig. 9). These considerations suggest that the Cs^{137} concentration in the body potassium of man (if incorporated via the ecological cycle) would be about 3 per cent of the total Cs^{137} concentration in available soil potassium, i.e.

$$OR = \frac{(Cs^{137}/K)_b}{(Cs^{137}/K)_s} = 0.5(0.01 \times 2 \times 2) + 0.5(0.01 \times 2) = 0.03$$

If 1 mc/mile^2 gives 30 pc/g available soil potassium and the average surface deposition (December 1957) in the north temperate population belt was 36 mc/mile^2, the average Cs^{137} level in people (assuming entry through the ecological cycle) would be $30 \times 0.03 \times 36 = 32$ pc/g K, and people in the vininity of 40°N–45°N (Washington state, the Dakotas, Minnesota and Wisconsin) would average about 64 pc/g K.

Despite the broad generalizations in the above calculation, the values estimated from ecological considerations are surprisingly close to the measured values in people during the fall of 1957. The agreement, however, may be entirely coincidental and could result from direct fallout on vegetation, fortuitously making up for non-equilibrium of Cs^{137} with exchangeable soil potassium. We believe that Cs^{137}, at the present time, is entering the biosphere largely through direct deposition on vegetation because (a) its high fixation and slow leaching rate in soil make it unlikely that it can be in equilibrium with exchangeable soil potassium to the depth of the plant feeding zone, and (b) the Cs^{137}/K ratio in people and milk does not seem to be increasing in relation to integrated fallout.[13]

Whether Cs^{137} is entering the biosphere largely through direct fallout on vegetation or via the soil is very important. If entry is via direct contamination, Cs^{137} levels in people will come into equilibrium with the stratospheric fallout rate, and continuation of weapons tests indefinitely at the past rate will produce relatively little increase in the Cs^{137} body burden of the population. If entry is via the soil, continued testing at the past rate may result in equilibrium levels in people (in about 100 years) that will be from ten to fifteen times the present levels.[2,30,31] Furthermore, immediate cessation of tests would, in the former case, result in an immediate decline in the Cs^{137} levels in people at a rate comparable to the half-time of stratospheric fallout. In the latter case, the

levels in people would continue to rise and reach a maximum in about 1965.[2]

DISCUSSION

Radiation dose from Cs^{137}

About 80 per cent of the world's population lives in the north temperate latitudes where the present (December 1957) average level of Cs^{137} deposition is estimated at about 36 mc/mile^2, and the average measured level in people is about 40 pc/g K. The weighted world population average would be only slightly less.

The external radiation dose rate from Cs^{137} surface deposition may be estimated from the following expression:

$$\text{Dose rate (mr/year)} = 5 \times 10^{-2} \times C \times E$$

where C is the deposition level in mc/mile^2, and E is the Cs^{137} γ-ray energy (0.66 MeV). The 1957 Cs^{137} deposition level of 36 mc/mile^2 gives a dose rate of 1.2 mr/year. The dose rate from past tests will increase by about 20 per cent* (from continued stratospheric fallout), reaching a maximum value of about 1.4 mr/year.[2] The infinite dose from 60 MT of fission (assuming about 8 years at equilibrium to compensate for dose during build-up) will be 67 mr, or about 1 mr/MT. This estimate unrealistically assumes no shielding by buildings and dwellings and no loss of Cs^{137} from the surface except through radioactive decay. It is virtually impossible to consider all parameters involved in a realistic estimation of an average external population dose from fission product surface deposition. STEWART *et al.*[9] have assumed an overall attenuation factor of 21. If a conservative factor of 10 is used, the infinite dose (assuming no further tests) is 7 mr. If tests continue at the rate of 10 MT of fission per year, the 30 year Cs^{137} external genetic dose may be about 30 mr (0.1 (10 MT/year $\times$ 1 mr/MT) 30 years), or about 1 per cent of the genetic dose from natural background.

In the event of continued testing, external γ-radiation from tropospheric and stratospheric deposition of other radionuclides, such as Zr^{95}, Ru^{106} and Ce^{144}, may be more important than that from Cs^{137}. The presence of these isotopes in the environment as a result of current testing has been reported.[32]

The internal radiation dose rate from present levels of Cs^{137} in people may be estimated from the following expression:[10]

$$q = \frac{2.8 \times 10^{-3} mW}{f_2 \Sigma E(\text{r.b.e.}) N}$$

in which q is the body burden in μc, m is the weight of the critical organ (in this case, assumed to be 70,000 g), f_2 is the fraction of the total body burden that is in the critical organ (1.0), $\Sigma E(\text{r.b.e.})N$ is an average energy term weighted for the relative biological effectiveness and absorption in the critical tissue (0.59), and W is the dose rate in rem/week.

A 70 kg man contains 139 g of body potassium;[13] therefore, a Cs^{137} concentration of 40 pc/g K gives a value for q of 0.0056 μc. Substitution in the above equation gives an internal total dose rate of 0.87 mr/year from the present average level of Cs^{137} in the majority of the world population.

If present levels in people are the result of ecological integration, the Cs^{137} internal dose rate (if weapons tests are stopped) may continue increasing to a maximum of about 1 mr/year in about 1965. Assuming Cs^{137} is removed from the soil only by radioactive decay, its mean time in the ecological environment will be 40 years, and the integrated infinite dose (plus 8 mr to correct for exposure received during build-up to equilibrium) from 60 MT equivalents of fission detonated to date will be about 48 mr, or about 0.8 mr/MT. At equilibrium with a given testing rate, the integral dose will equal MT/year $\times$ mr/MT $\times$ T, where T is the integration time. The average genetic dose (to age 30) from internally deposited Cs^{137} in equilibrium with a test rate of 10 MT of fission per year will be about 240 mr (assuming the dose to the gonads is the same as that to the total body). The internal plus external genetic dose will be about 270 mr, or about 10 per cent of the natural background dose.[33,34] The average 70 year Cs^{137} internal dose to the bone marrow (assuming it to be 80 per cent of the

* The preferential fallout model of MACHTA and LIST[5] predicts an increase of about 60 per cent. In this case, the predictions given in this report may be low by about 35 per cent.

total body dose) will be about 450 mr. The total internal plus external dose will be about 510 mr, which is approximately 5 per cent of the 70 year marrow dose from natural background. If, however, biospheric entry of Cs^{137} is largely via direct contamination of vegetation, the internal body burden should be in equilibrium with the rate of stratospheric fallout.

Assuming the present internal dose rate of 0.87 mr/year to be a result of equilibrium with the fallout rate (mean time equal to 6 years), the infinite Cs^{137} internal dose from 60 MT of fission, including 4 years at equilibrium (Fig. 1), will be about 8.7 mr, or about 0.14 mr/MT. The dose per megaton can be derived also from the estimated stratospheric inventory. With a stratospheric inventory of 23 MT (Fig. 1) and a fallout mean time of 6 years, the fallout rate is 17 per cent or 3.9 MT equivalents per year. Assuming the present Cs^{137} internal dose rate of 0.87 mr/year to be a result of equilibrium with this fallout rate, the integrated dose will be about 0.22 mr/MT equivalent of stratospheric fallout, or 0.13 mr/MT of fission detonated (assuming 60 per cent stratospheric injection). On the basis of this value, the average internal 30- and 70-year integrated genetic and bone marrow doses (assuming continued testing at the rate of 10 MT of fission per year) may be about 40 and 70 mr, respectively. The respective external doses, of course, would be the same regardless of whether Cs^{137} is entering the food chain via ecological integration or by direct contamination of vegetation. In the latter case, the total (internal plus external) 30 and 70 year doses may be about 70 and 130 mr, respectively. If weapons tests had stopped in 1957, all values would be much lower regardless of the mode of biospheric entry. External, internal, and total 30 year genetic and 70 year mean marrow dosage estimates under the various conditions are summarized in Table 6.

Preferential fallout in the north temperate latitudes[5,6] may result in radiation doses to the majority of the world population that will be about 50 per cent higher than those given in the table. The values for continued testing are based also on the assumption of a constant test rate of 10 MT of fission per year. At equilibrium, the doses will be directly proportional to the average test rate.

It is important also to keep in mind that these considerations have involved general averages only. The observed Cs^{137} levels in people showed a normal distribution with a standard deviation of about 36 per cent. Sufficient data are available to establish the normality of the curve to 3σ. If the distribution continues to be normal, only one person per 1000 would be expected to exceed 3σ or twice the average value. The probability of isolated individuals or local areas having still higher cesium levels than predicted from a normal distribution can be evaluated only by statistical treatment of

Table 6. *Estimated* 30 *year genetic and* 70 *year mean bone marrow doses from* Cs^{137} *biospheric contamination*

Conditions	30 year genetic dose (mrad)*			70 year marrow dose (mrad)†		
	External	Internal	Total	External	Internal	Total
Soil integration						
Tests stop	3	29	32	5	33	38
Tests continue	30	240	270	56	450	510
Direct uptake						
Tests stop	3	9	12	5	7	12
Tests continue	30	39	69	56	73	130

* Assuming dose to the gonads is the same as the whole body dose.
† Assuming marrow dose is 80 per cent of the whole body dose.

large amounts of data collected over a period of several years.

Significance of Cs^{137} *levels in the biosphere*

Regardless of the uncertainties inherent in the above estimations of radiation dose from Cs^{137} fallout, they are much more reliable than any biological interpretation of their significance to man's health and well-being.

Genetic response to radiation is generally believed to be non-threshold in character, and the dose required to double the natural mutation rate has been estimated as 30–50 r.[35,36] If Cs^{137} is entering the biosphere through soil integration and plant uptake, the total (internal plus external) radiation dose (~30 mr) from weapons tests to date may increase the incidence of mutations by about 0.05–0.1 per cent, and continued testing at the rate of 10 MT of fission per year (total 30 year dose ~270 mr) may increase the incidence by about 0.5–1 per cent. If Cs^{137} is entering the biosphere largely via direct fallout on vegetation, the total dose (and the increase in mutation rate) may be lower by a factor of about 4.

Present knowledge is not adequate to provide a basis for quantitative estimates of the potential somatic hazards of increasing background exposure from fallout. If, however, estimates of the present Cs^{137} and Sr^{90} levels in man are accepted, a crude indication of their relative importance is possible. The 1957 average maximum equilibrium Sr^{90} bone level in the world population, estimated from current bone analyses[37,38] was about 1.5 pc/g Ca.[1,2] This level may increase (from fallout of fission products still in the stratosphere) eventually reaching an average equilibrium value of about 1.8 pc/g Ca as a result of detonation of about 60 MT of fission. Since 1 pc/g Ca delivers 2.8 mrads/year, 1.8 pc/g Ca will deliver an infinite bone dose of about 200 mrads, or 3.3 mrads/MT. Assuming the mean marrow dose is about 30 per cent of that to bone,[39] the 70 year Sr^{90} marrow dose from a continuing test rate of 10 MT of fission per year would be about 0.7 rad (i.e. 0.3 × 0.0033 rad/MT × 10 MT/year × 70 years). The mean dose to the bone would, of course, be about 2.3 rads, or about 20–30 per cent of the average natural background dose.[1] The Sr^{90} mean marrow dose is about 1.4 times that estimated for Cs^{137}, assuming both are entering the biosphere via soil integration and plant uptake. If Sr^{90} is entering largely via the soil and Cs^{137} largely through direct contamination of vegetation, the 70 year marrow dose from Cs^{137} is about one-fifth that from Sr^{90}. Animal experiments with Sr^{89}[40] and clinical observations of radium poisoning[41–43], however, suggest that Sr^{90} may be a more likely osteosarcogenic agent than a leukemogenic one. In this case, a comparison of the potential somatic hazards of Cs^{137} and Sr^{90} fallout on the basis of their 70 year marrow doses is not valid.

The absolute somatic hazard of present and future levels of Cs^{137} and Sr^{90} fallout to world health depends critically on the shape of the dose–response curve. If radiation response shows a curvilinear relationship with dose, a factor (or factors) other than integrated dose must be important, and the biological effect may drop rapidly to insignificant levels at very low doses. Although the existence of an absolute threshold may never be proved empirically,[44] the existence of a practical one may be inferred from a curvilinear dose–response relationship. If, however, the dose–response curve is linear and passes through the origin, any amount of radiation (no matter how small) carries a certain probability of damage and a "single hit" cause and effect relationship, independent of dose rate and other factors, is required. In this case, a fraction of the natural population incidence of all somatic diseases produced by radiation must be attributable to natural background.

Lewis[45] and Court-Brown and Doll[46] have advanced the hypothesis that the leukemogenic response of man to low doses of radiation is linear and that any amount of radiation carries some probability of risk. Lewis' conclusions were based largely on reports of leukemia incidence in the Japanese following the bombings of Hiroshima and Nagasaki, and Court-Brown and Doll's were based on studies of several hundred cases of X-ray treated spondylitis. The latter authors suggested also that a radiation dose of from 30 to 50 r to the red bone marrow may double the leukemia rate

and that the probability of developing leukemia per year lies between 1×10^{-6} and 2×10^{-6}/r.

The linear hypothesis of somatic radiation damage has been questioned recently by MOLE,[44,47] Brues,[48] and FINKEL.[49] MOLE[44] pointed out the presence of a systematic error (the size of which becomes greater with increasing dose) in the mean population dose estimates to the Japanese which, if corrected, makes the dose–response relationship more likely curvilinear, and COURT-BROWN himself,[50] BRUES,[48] and MOLE[44] have pointed out that the data from the spondylitic cases are compatible with a marked curvilinear dose–response relationship if unselected for those cases receiving spinal irradiation only. Experimental data on animals also favor a curvilinear dose–response relationship,[48] and MOLE[47] has reported preliminary data which suggest that radiation-induced leukemia in mice may be dose-rate dependent. He further pointed out that the applicability of the data collected from the Japanese survivors and the spondylitic patients (in which the dose rates were of the order a million times greater than that from fallout) to the prediction of the leukemia hazard from slightly increased background depends on whether dose rate is a relevant parameter.

If, however, the non-threshold hypothesis and a leukemia doubling dose of 30 to 50 r are accepted in the interest of conservatism, the potential leukemia risk to the population from Cs^{137} and Sr^{90} fallout may be inferred. If Cs^{137} is entering the biosphere through soil integration and ecological uptake, the internal plus external 70 year marrow dose ($\sim$40 mr) from weapons tests through 1957 may increase the incidence of leukemia by about 0.1 per cent, or by about 150 cases per year, in the entire world population of 2.7 billion. A comparable number of cases may be expected from Sr^{90}. In this case, Cs^{137} and Sr^{90} fallout from a continuation of weapons tests indefinitely at a rate of 10 MT of fission yield per year may increase the world leukemia incidence on the order of 3000 to 6000 cases per year. In the event Cs^{137} is entering the biosphere by direct contamination of vegetation, its contribution to the leukemia incidence may be lower by a factor of about 4.

It should be emphasized that the above prediction appears to have no more basis, in the light of present knowledge, than does a prediction based on the assumption of a threshold dose–response relationship. In the latter case, one may be equally justified in predicting no increase in leukemia incidence from present levels of fallout. Such "armchair" predictions by well meaning individuals have been criticized recently.[49] The only reason for presenting such is to give the alternative possibilities in a situation for which there is no satisfactory answer at present and for which there may not be an unequivocal answer for many years. Regardless of which alternative turns out to be the case, present levels of Cs^{137} and Sr^{90} fallout represent a very small potential hazard compared to that of many other factors inherent in our complex society. Fortunately, the present rate of build-up of biospheric contamination is slow, allowing time for continued intensive and extensive study.

The above considerations probably provide little quantitative information on the potential threat of nuclear weapons tests to man's health and well-being. They do suggest, however, that Cs^{137} may contribute to the potential genetic effect of fallout, and whether it may be considered a relatively unimportant somatic hazard in comparison with Sr^{90} will depend on the nature of the radiation dose–response curve and on its route of entry into the biosphere.

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MEASUREMENT OF RADIOACTIVITY IN AIR FILTERS

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Abstract—Measurements of the fallout fission product activity that accumulates in the air filters employed in ventilation systems are described. A general method for determining the γ-activity of pelletized filter samples using a NaI well-type crystal spectrometer is discussed. The photofraction of a well-type crystal was measured and data are presented for γ-ray energies from 0.12 MeV to 1.12 MeV. It is suggested that building air filters be employed in monitoring programs connected with laboratories, reactors, and studies on fallout.

INTRODUCTION

THE accumulation of fallout fission product activity in the air filters employed in building ventilation systems suggests a sensitive and economic method of monitoring for air-borne particulates. It also suggests that there may be some cause for concern in the handling of used filters. We have observed relatively high activity in ventilation filters of different types and from different locations in North Carolina. Filters reading as high as 1 mr/hr near the surface as measured by a G.-M.-type survey meter have been found. It has been our experience that, barring accidents, much less activity is observed in the fume hood filters of radioisotope laboratories using curie quantities of various isotopes.

Because of the large volume of air processed by building ventilation systems, the filters will concentrate a larger amount of the air activity than would be collected by the usual air-filter monitor. Air-filter samplers that are commonly used have air flow rates from 1 to 50 ft^3/min while building ventilation systems may employ several blowers rated at over 10,000 ft^3/min each.

The utilization of building air-filters could introduce a substantial savings in monitoring programs, particularly where off-site monitoring is necessary or desirable. Also, it would appear that valuable information concerning world-wide fallout could be obtained by studying the activity in building-filters.

Since fission products from fallout also concentrate in air filters in reactor buildings, it seems that it is almost mandatory that both off-site and on-site building filters be studied in order to be sure that the activity is not coming from the reactor.

In this paper we describe some of the measurements made over a period of about 1 year of the activities present in both intake and exhaust filters taken from buildings in North Carolina.

METHOD OF MEASUREMENT

Because of the large volume of filter material plus collected dust, it was decided that γ-ray measurements would produce the most significant results. Samples were prepared by pelletizing approximately 10 g of filter material (either paper or glass wool) to form a cylindrical pellet $\frac{1}{2}$ in. dia. $\times$ 1 in. long. Paper filters can be ashed before pelletizing and thus further increase the concentration of activity. The samples were then counted in a NaI well-type scintillation counter. A comparison of the activities of burned and unburned samples showed that the loss of activity by ashing was negligible. Gamma-ray spectrum measurements were made using a single channel pulse height analyzer with a standard NaI well-type crystal (Harshaw type 7F8).*

* The Harshaw type 7F8 well-type crystal is 1.75 in. dia. $\times$ 2 in. high. It has a 0.75 in. dia. well, 1.5 in. deep.

In order to determine activities it was necessary to first determine the contribution of each γ-ray energy to the total counting rate so that correct efficiency factors could be applied. This was done by the method of photofractions. For monoenergetic gammas, the photofraction is defined as the ratio of the number of photons totally absorbed to the total number of photons undergoing interactions in the crystal. It is represented by the ratio of the area under the total energy peak to the area under the total pulse height distribution. BERGER and DOGGETT[1] have made photofraction calculations by the Monte Carlo method for solid crystals of various sizes for energies of 0.279 to 4.45 MeV. MILLER *et al.*[2] made more refined calculations taking into account the probability of escape of the annihilation radiation. The photofraction of a well-type crystal has not been treated theoretically and has therefore been measured in this laboratory. A comparison is made between the integral count taken of the total energy peak and the total integral count extrapolated to zero energy. Results are plotted in Fig. 1.

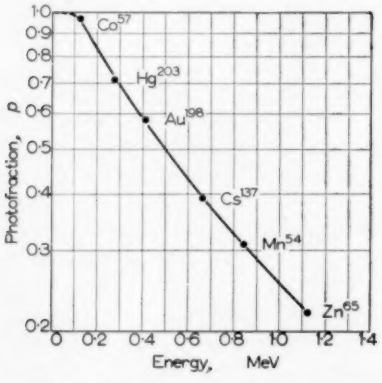

FIG. 1. Photofraction (p) vs. γ-ray energy for NaI well-type crystals.

These data were used to determine the contribution of each energy peak to the total integral count. Two or more peaks are done by subfractions.

The efficiency of the standard well-type crystal has been measured by BASKIN *et al.*[3] for several isotopes. An efficiency measurement for a similar though not identical crystal is reported in ORNL Progress Report 1160.[4] This curve was used together with a calculated curve to extend the data of BASKIN to other energies. The efficiency data of WOLICKI *et al.*[5] were used to calculate an efficiency curve for two solid crystals having diameters 1 in. and a thickness of $\frac{1}{2}$ in. with a source sandwiched between the two crystals on the axis of the two crystals. Correction was made for escape of photons from the well. This geometry should be nearly equivalent to that of the Harshaw crystal. The efficiency curve of the Harshaw crystal is then drawn through the measured points of BASKIN with the two adjacent curves serving as a guide regarding the shape. These curves are presented in Fig. 2.

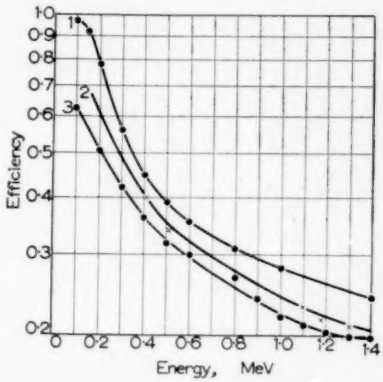

FIG. 2. Efficiency vs. γ-ray energy for NaI well-type crystal. Curve 1—WOLICKI *et al.*[5]; Curve 2—BASKIN *et al.*[3]; Curve 3—BORKOWSKI.[4]

RESULTS

Fig. 3 shows the γ-ray spectrum of a building air filter sample taken in September, 1957. Plotted on the same graph is the spectrum of a sample of reactor fuel solution made approximately 2 days after the reactor was shut down. The previous history of the reactor operation was one of intermittent low level operation over a period of 3 months. The high energy peak corresponds to 0.72–0.75 MeV and can be attributed to the mass 95 isotopes, Zr^{95}–Nb^{95}. The second peak is due to Ru^{103} with possible contributions from Nd^{147}. The low energy peak appears to be Ce^{141}. Measurements made of

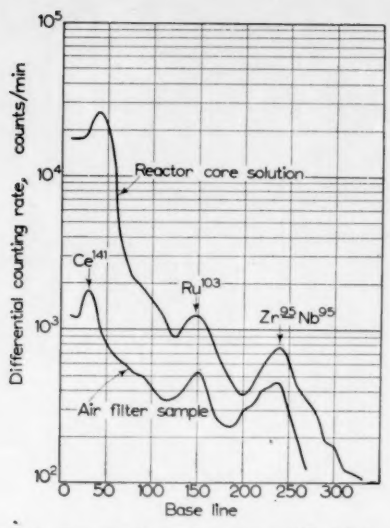

FIG. 3. Comparison of spectra of Raleigh reactor core solution and a building air filter sample.

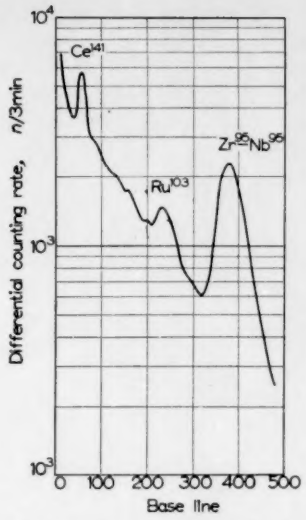

FIG. 4. Gamma-ray spectrum of a building air filter sample.

the decay of the sample support these conclusions. After 10 months decay the filter sample exhibits an almost pure Cs^{137} spectrum, as would be expected.[6]

Essentially the same spectra are obtained from samples collected from different locations at the same time. However, different spectra are obtained from filter samples taken at different times. The spectra in Fig. 3 show approximately the same amount of the 0.5 MeV and the 0.72–0.75 MeV activities indicating 10–30 day old fission products,[7] commensurate with the weapons testing of the summer of 1957. The spectrum from a filter sample taken during early 1958 is shown in Fig.4, which shows the 0.72–0.75 MeV peaks predominating. This would be the case if the fission products were about 6 months old[6,7] and is again in keeping with the bomb testing.

The Zr^{95}–Nb^{95} activity in the sample of Fig. 4 was 0.05 μc and the Ce^{141} activity was 0.004 μc as determined by the method given above. Using the known filter area and air flow rates, the average specific air activity is calculated to be 2.2×10^{-12} $\mu c/cm^3$, assuming a filter collection efficiency of 90 per cent. A continuously recording monitor using an end-window G.-M. tube was mounted over the filters during the collection period. This monitor showed a very gradual rise in the activity during this period. Therefore, we regard the figure given above for the specific air activity to be a good order of magnitude for existing conditions; that is, it was evident from the continuous monitor that there was no large changes in air activity during the period the sample was taken. Efficiencies of different types of building filters can be determined by comparing the activity in the air on opposite sides of the filter in question. Standard air monitoring techniques are used for this comparison.

CONCLUSIONS

From our experience, the usefulness of building ventilation air filters in monitoring for air-borne particulate activity seems to be well suited to γ-emitting isotopes with half-lives longer than a few days. While this method does not provide direct information about pure β-emitting isotopes such as Sr^{90}, particular β-activities can be inferred from the known fission yields and the measured γ-activities. There is no apparent reason why continuous monitors could not be devised for use with building filters in monitoring for both α and β as well as γ-emitting isotopes.

Although the inhalation hazard from radioactive dusts, that are stirred up when building air filters are changed or cleaned, cannot be generally specified, it would appear that a hazard could exist during such operations and that appropriate precautions should be taken.

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RETENTION AND LOCALIZATION OF PHOSPHORUS-32 IN RAT OVARY*

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Abstract—Data for retention of P^{32} in ovary and bone over a period of 56 days following single intraperitoneal administration were obtained. Concentrations of P^{32} in ovary decreased as a power function with time. Concentrations in bone remained constant throughout the experimental period. Autoradiograms showed that P^{32} was heterogeneously localized in ovary tissue. Application of these findings to the establishment of maximum permissible concentrations is discussed.

RADIATIONS from both external and internal sources may have deleterious effects upon ovarian tissue.[1] In order to facilitate evaluation of the potential hazard of P^{32} deposited within the ovary its localization was studied by autoradiographic techniques. These results together with radiochemical assays of ovarian P^{32} concentrations were compared with the P^{32} concentrations in bone. From these comparisons it appears quite possible that considerations of genetic damage may ultimately prove to be the limiting factor in determining maximum permissible limits for P^{32}.

EXPERIMENTAL

A randomly selected group of 65 young adult female Sprague–Dawley rats (wt. range: 175–256 g) were intraperitoneally administered P^{32}, as $Na_2HP^{32}O_4$, at dosages ranging from 0.5 to 10 μc/g of body weight. Following injection the rats were killed at time intervals of from one to 56 days. One ovary from each animal was removed and fixed in 10% neutral formalin, dehydrated with cellosolve and benzene and embedded in tissue mat. Adjoining pairs of sections, 6 μ thick, were selected for histological and autoradiographic processing. One section was stained routinely with hematoxylin and eosin for orientation. The other was exposed at 4°C for 8 days on non-screen X-ray film, plus a "*T*" coat. After development with Eastman X-ray developer at 20°C the emulsion slides were washed and cleared in Eastman Kodak acid fix for 30 min at room temperature. The slides were washed in running water 1 hr to remove the fix and stained with hematoxylin and eosin.

The second ovary, one femur, and the pelvis were removed from each animal, wet ashed with concentrated HNO_3 and diluted to known volumes. The concentrations of P^{32} in suitable aliquots of these samples were measured with appropriate corrections for self-absorption and radioactive decay.

RESULTS

1. *Autoradiographic localization of* P^{32} *in ovary*

It was necessary to vary exposure time and development in order to obtain a satisfactory autoradiographic image. Ovaries of animals killed one day after receiving 2.5 μc P^{32} per g body wt. and ovaries of animals killed 8 days after receiving 10 μc P^{32} per g body wt, gave comparable autoradiograms on the basis of visible density variation. Fig. 1(a) is an autoradiogram which illustrates localization of P^{32} 1 day after injection. Fig. 1(b) is an adjacent histologic section. Conclusions which may be drawn from the autoradiogram are entirely

* This paper is based on work performed for the Atomic Energy Commission under contract W-31-109-Eng-52.

topographic and are not for quantification. The autoradiogram of Fig. 1(a) shows that the Graafian follicles markedly concentrated P^{32} in the granulosa layer. The apparent grain density was greatest over the cellular elements lining the follicular cavity and those surrounding the ova. Grain density over the fluid area of the follicles was negligible. Atretic and degenerating follicles showed only a slight uptake of P^{32} in the walls and follicular fluid. Some of the *corpora lutea* definitely concentrated P^{32}, but in others only small amounts were observed. The vascular system did not contain appreciable quantities of P^{32}. Concentrations of P^{32} in the interstitial tissue of the ovary were moderate after 1 day, but were slightly greater in ovaries of animals killed 8 days after injection.

2. *Deposition of* P^{32} *in bone*

Concentrations of P^{32} in femur and pelvis of the animals at several time intervals following its intraperitoneal administration are shown in Table 1. There was no statistical evidence of any direct relationship between percentage P^{32} deposition and dose level. Corrected for radioactive decay the concentrations of P^{32} did not show any statistically valid trend to either increase or decrease throughout the experimental period of 56 days. The weighted average concentrations of P^{32} and their standard deviations in femur and pelvis were 1.44 ± 0.12 and 1.59 ± 0.12 per cent of administered dose per g, respectively.

Table 1. *Phosphorus* (P^{32}) *in femur and pelvis following intraperitoneal injection*

Time (days)	Number of animals*	Mean % P^{32}/g ± s.d.†	
		Femur	Pelvis
1	9	1.38 ± 0.13	1.48 ± 0.13
7	6	3.65 ± 2.27	4.62 ± 3.42
8	13	1.28 ± 0.69	1.41 ± 0.74
13	3	3.08 ± 1.17	3.16 ± 0.60
14	4	2.58 ± 0.95	2.45 ± 0.82
28	8	2.37 ± 1.31	2.70 ± 1.46
42	6	2.91 ± 1.28	3.30 ± 1.69
56	3	1.40 ± 1.19	1.64 ± 1.53
Weighted average		1.44 ± 0.12	1.59 ± 0.12

* Twelve animals dosed with 10 μc P^{32}/g body wt. succumbed shortly after injection.

† Radiophosphorus values obtained from surviving animals showed no statistical relationship to dosage level of P^{32}. All values are combined means from randomly dosed animals. The average femur weight was 0.574 g.

Table 2. *Phosphorus* (P^{32}) *in ovary following intraperitoneal injection*

Time (days)	Number of animals	Mean % P^{32}/g ± s.d.*
1	10	0.800 ± 0.100
7	6	0.447 ± 0.266
8	13	0.301 ± 0.300
10	1	0.068
13	3	0.454 ± 0.136
14	3	0.164 ± 0.019
28	8	0.075 ± 0.029
40	1	0.064
42	6	0.047 ± 0.014
56	3	0.086 ± 0.135

* The average single ovary weight was 0.042 g.

3. *Retention of* P^{32} *in ovary*

The concentrations of P^{32} in ovary for the several experimental time intervals are shown in Table 2 and are plotted on a logarithmic scale in Fig. 2. This figure shows that the retention of P^{32} by ovary may be characterized by a power function in terms of the fraction retained at a given time after administration.

$$R = 1.25\, t^{-0.76} \qquad 1 \leq t \leq 56$$

where R is per cent of administered dose retained per gram after t days, and 1.25 represents the retention at the end of 1 day. More than 50 per cent of the ovarian P^{32} was lost by the third day, and more than 90 per cent by the twenty-fifth day following administration.

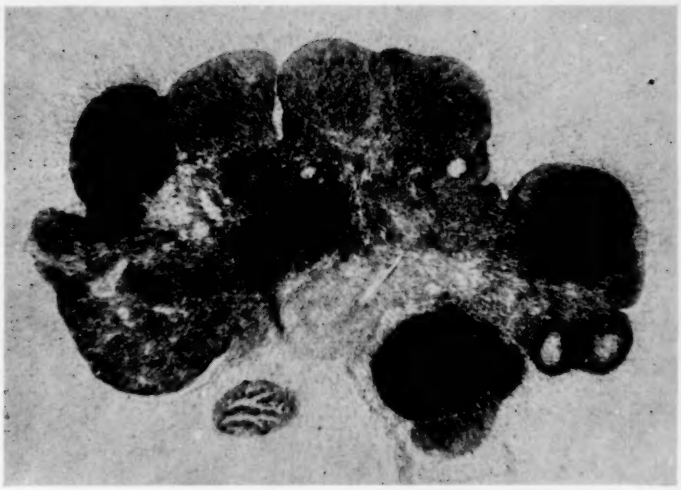

FIG. 1(a). Autoradiogram of ovary preparation following administration of P^{32}. P^{32} concentrated around follicles and in *corpora lutea*. Animal sacrificed after 24 hr.

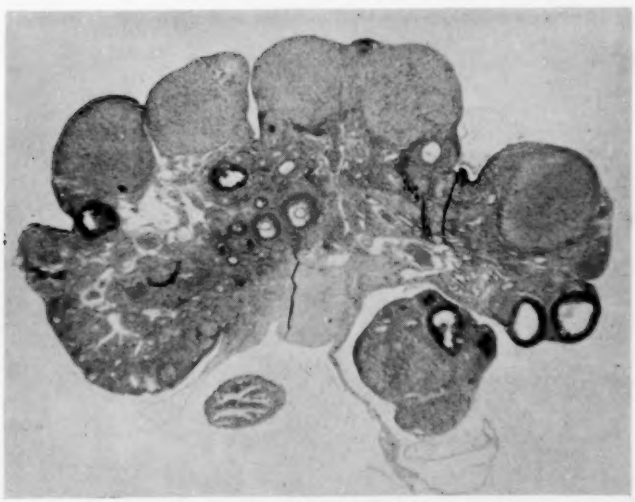

FIG. 1(b). Stained ovary preparation following administration of P^{32}.

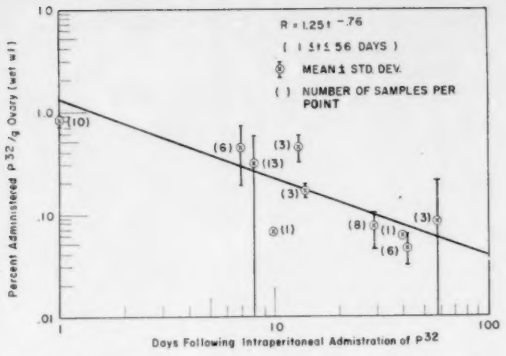
R = 1.25 t^-.76
(1 ≤ t ≤ 56 DAYS)
MEAN ± STD. DEV.
( ) NUMBER OF SAMPLES PER POINT
Percent Administered P 32 /g Ovary (wet wt)
Days Following Intraperitoneal Admistration of P32

FIG. 2. Retention of phosphorus (P^{32}) in rat ovary.

DISCUSSION

The marked concentration of P^{32} in the granulosa layer lining the follicular cavity and in the corpora lutea, and the small to moderate amounts of P^{32} in other regions of the ovary agree with results obtained in autoradiograms of mouse ovary[2] and rabbit ovary 2–6 hr after P^{32} administration.[3] With other autoradiographic studies these findings suggest close relationships between growing tissue and enhanced concentrations of P^{32} and may be explained by an increased affinity of growing tissue for P^{32}.[4,5] The autoradiogram thus indicates a heterogeneous localization of P^{32} within the ovary.

The spatial relationship of the autoradiogram to the whole organ is important in calculation of the radiation dosage. The autoradiogram records only a thin cross-section of ionization paths of radioactive particles emanating from a thin tissue section. In so far as the ovary consists of many similar juxtaposed tissue sections the corresponding microconcentrations of P^{32} are randomly dispersed throughout its volume. Tentative conclusions based on the gross concentration data, where a uniform concentration of P^{32} is assumed, may be modified by this heterogeneous spatial arrangement. Calculated on the basis of physical half-life and the power function which expresses ovarian retention, the long term continuous administration of P^{32} will result in an ovary concentration of 10 per cent of a single day's dose per gram.[6]

In bone the absence of any appreciable turnover of P^{32} indicates that its accumulation will be limited only by its physical half-life. On this basis the experimental value of 1.5 per cent per gram following intraperitoneal injection indicates an eventual accumulation with continuous oral administration of about 23 per cent of a single day's dose per gram. Assuming uniform distribution of P^{32} in bone as well as in ovary, the radiation dose to ovary will be over one-third as great as that to bone. Rather than the present 560 mrads obtained by comparison with radium,[7] comparison of a maximum permissible weekly rate of 300 mrads to bone with the conservative permissible gonadal dose of 100 mrads[8] indicates that ovary may be as critical as bone in the rat. The possibility of heterogeneous distribution of P^{32} in bone as well as the experimentally observed non-uniform distribution of P^{32} in ovary could further alter this relationship. Additional consideration must be given to the relative radiosensitivity of tissue in the region of P^{32} microconcentrations. Some additional uncertainty in the applicability of small animal data to humans is found in the possibility of species differences in P^{32} retention characteristics, the disparities in relative organ sizes, and the possibility of differences in relative tissue radiosensitivity. These factors, which require much further investigation before a firm conclusion may be reached, do not exclude the possibility that the limiting factor in human permissible limits for P^{32} exposure may be genetic alteration rather than somatic damage.

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GAMMA RESPONSE AND ENERGY LOSSES IN THE ABSOLUTE FAST NEUTRON DOSIMETER

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Abstract—The γ-response and the associated question of the neutron energy lost under the bias needed to discriminate γ-radiation in the fast neutron dosimeter is considered for the cases where the γ-ray dose rate is high (>1 r/hr). The particular dosimeter considered is the so-called absolute fast neutron dosimeter which is calibrated by means of an internal α-source. Basic data are given for "effective" energies of 100 kVe and 200 kVe X-rays and Co^{60} γ-radiation. For very high γ-ray intensities (from 10 to 100 r/hr) a graphical method of treating the pulse height distribution data is suggested. Thus, for example, a dose rate of 1 mrad/hr of Po–Be neutrons can easily be determined even when the γ-dose rate is as high as 100 r/hr.

1. INTRODUCTION

A METHOD of fast neutron dosimetry which uses a proportional counter designed according to the requirements of the Bragg–Gray principle has been described elsewhere.[1,2] The feature of the method which permits the fast neutron tissue dose to be measured in absolute units and at the same time discriminate against γ-radiation has lead to extensive use of this type of dosimetry. In some applications it has been found that it is possible to use the method even when the γ-ray dose is as high as 100 r/hr. The purpose of this report is to discuss the performance of the instrument in the case where the γ-ray dose rate is very high, and to consider the associated question of the fraction of the neutron dose which is lost under the bias necessary for the discrimination of these γ-rays. If the γ-intensity is low, then essentially no bias is needed and hence the fraction, f, of the energy lost under the bias is negligible. In most applications even when the γ-dose rate is from 10 to 100 times the neutron dose rate, f is negligible. The proportional counter method of neutron dosimetry has recently been criticized by PETROV,[3] on the basis that excessive fractions of the neutron dose are lost under the bias. We surmise that he failed to note that an appreciable bias is needed only when the γ-dose rate is large. Furthermore, his calculations for the fraction lost under the bias appear to be too large as shown by RITCHIE.[4] This appears to be attributable to misconceptions on the exact function of the binary type pulse integrators[5] which are often used in this method.

Fig. 1 is shown to illustrate the problem of γ-response and energy loss. Curves are drawn which show the relative count rates for γ-rays, fast neutrons, and the 5.14 MeV α-calibration source. The neutron dose is proportional to the area under the neutron curve ($A_1 + A_2 + A_3$), and this may be measured directly in units of MeV/g of ethylene (hence tissue)[1] by means of the α-particle source. For example, in Fig. 1 the α-particle pulse height, V_0, is 190 V, thus 5.14 MeV = 190 V. The area A_1 represents the part of the neutron dose which is lost under the bias, B ($B = 5$ V in the case shown in Fig. 1). The rectangular area A_3 is not lost under the bias, but is a part of the energy dissipated by protons making pulses larger than the bias. The area A_4 represents the γ-response for the particular γ-intensity. Obviously for a given intensity increasing B makes A_4 smaller but A_1 larger.

Two methods are used to determine $A_1 + A_2 + A_3$: the direct graphical integration

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

method and the automatic pulse integration method. In the graphical method, naturally A_3 is included and even A_1 can be estimated. This will be further illustrated in Section III. The binary pulse integrators are designed to measure the area $A_2 + A_3$. In the case of the simple four-stage binary integrator,[2] it was shown that the integrator overestimates $A_2 + A_3$, thus partially compensating for the area A_1. The reason for showing A_3 is to emphasize that in no case is this part of the energy in question.

Basic data required for the determination of the fraction, f, of the energy lost as a function of neutron energy, E, and discriminator bias, B, have already been presented.[2] Values of f, defined as $f = A_1/(A_1 + A_2 + A_3)$ are shown in Table 1.

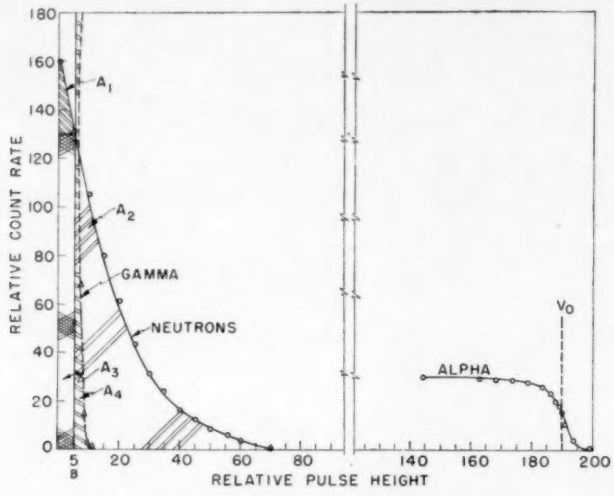

FIG. 1. Relative count rate vs. relative pulse height for neutrons, γ-rays, and α-particles in the absolute fast neutron dosimeter.

Table 1. Fraction $f \times 10^2$ of energy spent by recoils losing less than the bias energy B in the counter for various neutron energies

Bias		Fast neutron energies							
Energy (MeV)	PHS (V)	0.5 MeV	1.0 MeV	2.0 MeV	3.5 MeV	4.8 MeV	14 MeV	Po–B	Po–Be
0.074	2.6	8.9	2.0	1.5	1.3	1.4	2.5	0.6	1.4
0.14	5.3	19.5	9.4	4.1	3.5	2.8	8.5	2.8	4.5
0.21	7.8	32.0	12.9	7.6	5.6	5.9	16.2	6.5	10.1
0.28	10.5	52.6	23.5	12.3	9.1	9.8	25.8	9.9	14.4
0.36	13.2	73.0	33.3	18.8	12.8	16.2	36.9	15.2	20.5

II. DETERMINATION OF γ-RESPONSE

The irradiation of the counter was carried out by means of a X-ray machine and a 2.1c Co^{60} source. The electronic equipment used was an A1A preamp, A1D linear amplifier used in the 0.5 mc bandwidth, scaler with a 5 μsec resolution, and a stable high voltage supply.

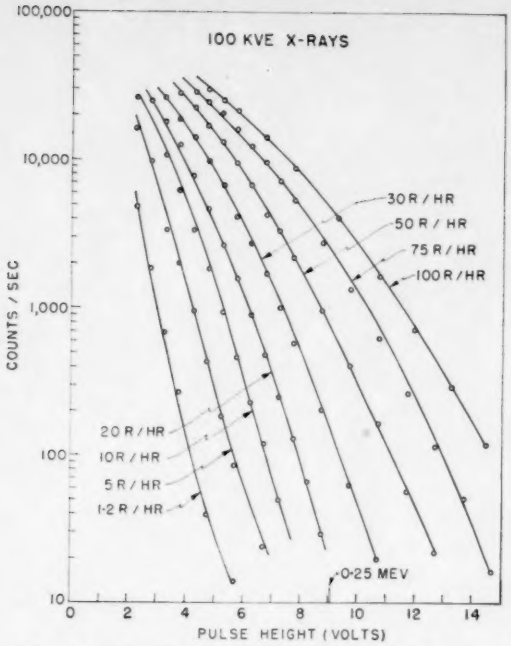

FIG. 2. Counts/sec vs. integral pulse height for 100 kVe X-rays.

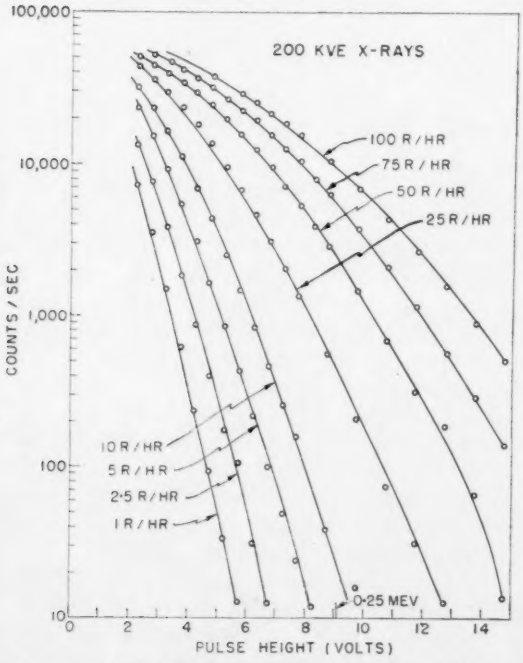

FIG. 3. Counts/sec vs. integral pulse height for 200 kVe X-rays.

With 100 kVe* X-rays (produced by heavily filtered 150 kVp X-rays), 200 kVe X-rays (produced by heavily filtered 250 kVp X-rays), and a Co^{60} γ-source, counts as a function of integral pulse height were determined for radiation intensities ranging from 1 to 100 r/hr.

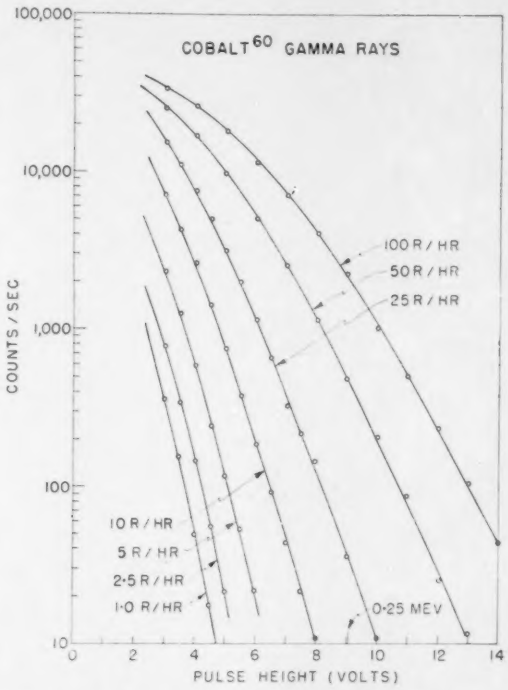

FIG. 4. Counts/sec vs. integral pulse height for Co^{60} γ-rays.

Using these data, families of curves (Figs. 2, 3 and 4) were plotted for the 100 kVe X-rays, 200 kVe X-rays, and Co^{60} γ-rays, showing counts per second as a function of pulse height.

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* kVe stands for "kilovolts effective" and is used here as unit of "effective" X-ray energy, as defined by L. S. TAYLOR, *Radiol.* **29,** 22 (1937). Al and Cu filters were used by H. H. HUBBELL, JR. (J. C. VILLFORTH, R. D. BIRKHOFF and H. H. HUBBELL, JR., *Comparison of Theoretical and Experimental Filtered X-ray Spectra* ORNL-2529, 1 July, 1958) to determine these effective energies as recommended by GLASSER, QUIMBY, TAYLOR and WEATHERWAX, *Physical Foundations of Radiology* p. 244. Hoeber, New York (1952).

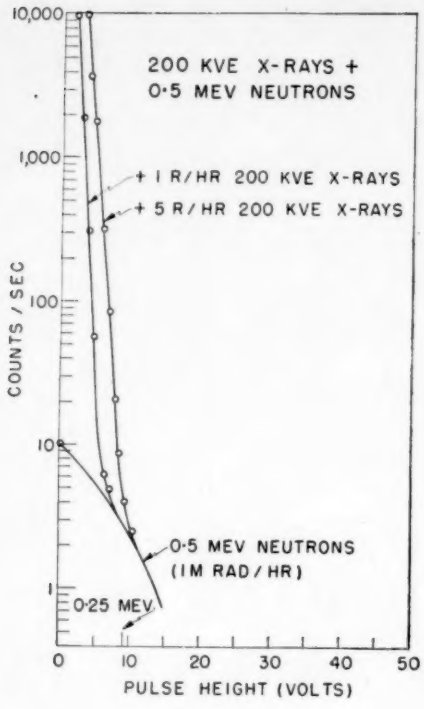

FIG. 5(a). Counts/sec vs. integral pulse height for mixtures of 200 kVe X-rays with 0.5 MeV neutrons (1 mrad/hr).

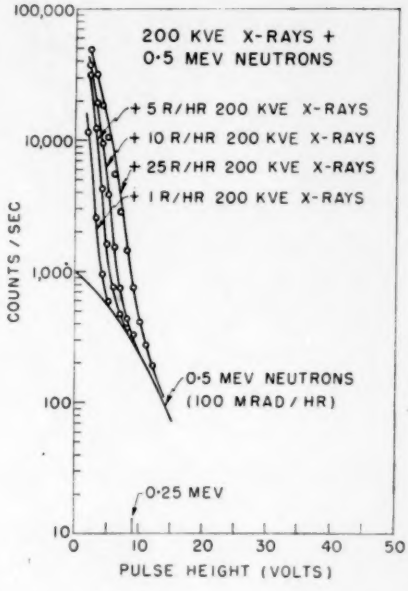

FIG. 5

FIG. 5(b). Counts/sec vs. integral pulse height for mixtures of 200 kVe X-rays with 0.5 MeV neutrons (100 mrad/hr).

III. RESULTS FOR MIXTURES OF NEUTRONS AND γ-RAYS

In this section the results for mixtures of neutrons of various energies with X- or γ-radiation will be considered. The neutron data from reference (2) will be combined* with the X- and γ-ray data in Section II. Fig. 5(a) shows the most severe case, i.e. the combination of 0.5 MeV neutrons with 200 kVe X-rays. In Fig. 5(a) the neutron intensity corresponds to 1 mrad/hr, and the γ-ray intensity for 1 r/hr and 5 r/hr. With 1 r/hr the γ-response becomes appreciable for $B \leqq 0.20$ MeV, hence the fraction of the dose, f, which must be estimated by extrapolation is 32 per cent (see Table 1). At 5 r/hr the bias energy is about 0.28 MeV and 53 per cent of the dose must be estimated. When the neutron dose is higher, a higher γ-intensity may be tolerated [see Fig. 5(b)]. In limiting cases of this type it is recommended that the graphical method of determining the area under the neutron curve be used. In practice, low energy X-rays could be attenuated with high-Z materials without appreciable effect on the neutron dose. The above example, however, serves to define the limits of the proportional counter method for separating the neutrons from γ-radiation.

Fig. 6 shows a more typical case, i.e. the Po–Be neutron spectrum mixed with Co^{60} γ-radiation. Again, in this case the neutron dose rate is 1 mrad/hr, but the γ-dose rate ranges from 1 r/hr to 100 r/hr. Reference to Fig. 3 and Table 1 shows that the fractions, f, which

* In an independent experiment the combination procedure was validated, i.e. γ-intensities as high as 100 r/hr did not affect the pulse distributions due to fast neutrons or the α-calibration source.

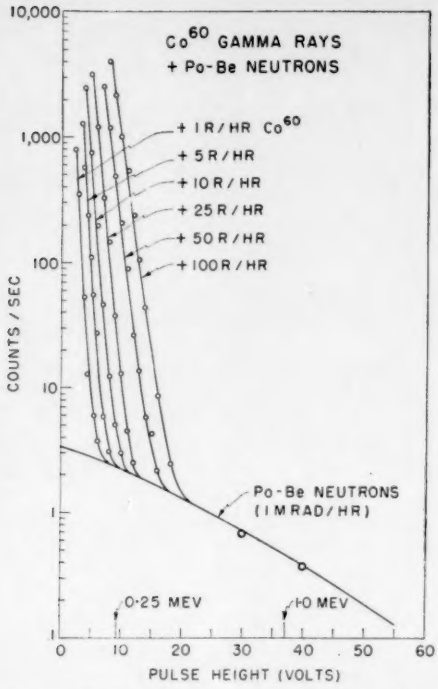

FIG. 6. Counts/sec vs. integral pulse height for mixtures of Co^{60} γ-rays with Po–Be neutrons.

require estimation by extrapolation are approximately 10 per cent, 12 per cent, 14 per cent, and 18 per cent for Co^{60} intensities corresponding to 1 r/hr, 5 r/hr, 10 r/hr, and 25 r/hr, respectively.

The graphical method for determining the amount of energy lost under the bias involves: (1) plotting the log of total count rate, due to neutrons plus γ-radiation, against the integral discriminator level (as in Fig. 6); (2) noting the point where γ-radiation causes a sharp rise in the curve; (3) extrapolating the neutron curve for pulse level below this point; and (4) transferring the entire neutron curve to a linear scale and determining the full area under this curve.

The graphical method outlined above is necessary for precise measurement only when the γ-radiation dose rate is very high (from 10 to 100 r/hr), and when the fast neutron dose rate is low ($<$100 mrad/hr). In other cases the binary pulse integrators give adequate accuracy, and require less time and effort.

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STRONTIUM-90 DISTRIBUTION IN THE HUMAN SKELETON*

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Abstract—Strontium-90 is now present in the skeletons of all living persons throughout the world as a result of nuclear detonations. Since this isotope is nonuniformly distributed within the adult skeleton, it is necessary to define this distribution in order to more adequately estimate the resultant radiation hazard as well as to evaluate and compare data from a variety of bones of different individuals.

The principal bones from many cadavers were analyzed. While some skeletons were assayed very extensively, the bulk of the investigation centered on rib, vertebrae and long bone shaft. In the case of adults, an aliquot of the total body skeleton was also analyzed in each case.

In adults the strontium-90 concentration per gram calcium in vertebrae, rib and long bone shaft is 1.8, 1.1 and 0.5 times skeletal average respectively. In newborns and young children, the distribution is much more uniform although these data are more limited.

These findings together with the fact that stable strontium is uniformly distributed throughout the adult skeleton show that the nonuniformity of strontium-90 is a temporal function of changing diet. If radiation damage from strontium-90 is a threshold phenomena, the distribution is of particular importance since the highest local concentration areas will first exceed the threshold. If the damage is nonthreshold in character, the distribution is still of some significance since some areas (e.g. bone marrow) are presumably more susceptible to radiation damage than other areas.

INTRODUCTION

STRONTIUM-90 originating from nuclear detonations has now been disseminated throughout the globe so that measurable quantities are present in practically all surface soil, food and human bone. The biological chain of strontium-90 transfer is under extensive investigation so as to assess the present and potential hazard.[1,2,3] The ultimate quantity of interest is, of course, the amount of strontium-90 which reaches the human skeleton. The distribution within the skeleton is also important for the proper evaluation of the physiological hazard. Thus, if the areas of the skeleton which contain a higher than average strontium-90 concentration are those which are more susceptible to radiation damage, the strontium-90 in the skeleton will be more hazardous than if it were distributed uniformly.

A further practical consideration which has made the determination of the strontium-90 distribution within the skeleton imperative has been the need for properly computing the skeletal load of this radioisotope in man from analysis of single bones. For the past 5 years the Lamont Laboratory has been assaying bones received from a world-wide network.[3] Individual bones from autopsy are supplied, e.g. vertebra, rib, femur and tibia. If valid comparison is to be made between bones of different types, the skeletal distribution must be known.

The results of SCHULERT *et al.*[4] on skeletal distribution in humans who had received a single tracer dose of strontium-85 show that the bones vary widely in the concentration of this isotope shortly after administration. Their average results at time intervals of from 1 day to 4 months show vertebra and rib concentrations as 6.0 and 1.9 times femur shaft, respectively. This clearly will not be the situation for young children who are depositing

* Lamont Geological Observatory Contribution No. 338.

new bone or with adults who have had a constant strontium-90 diet for a lifetime. This is evidenced by the fact that stable strontium is uniformly distributed throughout the skeleton.[5] The present study was undertaken to determine the actual strontium-90 distribution in the human skeleton at the present nonequilibrium state. It must, of course, lie between the distribution pattern of the single dose and uniformity.

PROCEDURE

Samples of the principal bones from adult cadavers were analyzed. Since death in most cases was due to a variety of natural causes rather than sudden accident, the results may not be entirely applicable to healthy adults. Effort was made to obtain a representative sample of each bone to typify as closely as possible samples obtained in the world-wide survey program. Therefore the bulk of the investigation centered on rib, vertebra, and long-bone shaft. In a concluding series equal amounts of identical bones from two groups of cadavers were pooled. This permitted much more accurate analysis.

Whereas the sampling of adult skeletons is adequate, that in children is much more limited. One complete skeleton of a 13 year old male was analyzed. In eight instances, from two to four bones each from young children were obtained for analysis. A number of fetal and neonatal skeletons were also measured.

The samples were prepared and assayed for strontium-90 using procedures reported elsewhere.[6,7] Briefly, the method consists of chemically isolating the alkaline-earths, allowing the yttrium-90 daughter of strontium-90 to accumulate, milking off and counting the yttrium-90. Owing to the relatively low activity of long-bone shafts, the sample required for precise assay is often too large to use this procedure conveniently. In many of these samples as well as total body samples, therefore, a preliminary separation of strontium from calcium by an adaptation of the method of Burgus[8] was performed.

RESULTS

The concentrations of strontium-90 in three bone types and the total body of twenty-seven individuals and three integrated sets of five individuals each who died in 1956 and 1957 are given in Table 1. In most cases a number of long bones were analyzed in a search for significant differences but within the experimental error these were not observed. The averages indicate the relative activity in vertebrae, rib and long bone. If only one or two bones of a given type were analyzed the error is the standard deviation of the counting statistics. In cases in which three or more bones were analyzed the average value is given together with the standard error on the mean. In some instances the counting error is quite large owing to the low concentration of strontium-90. The uncertainty indicated by these standard errors is dominated by the limitation in the precision of the radiochemical analysis rather than by biological variation.

The strontium-90 concentration in the children's bones is given in Table 2. Table 3 summarizes the relative strontium-90 concentrations in the important bones of adults, children and newborns.

Table 4 compares the present values with those used previously[3] based on preliminary data on the strontium-90 distribution in cadavers and short-term strontium-85 investigations.[4] The results from this more comprehensive comparison show that the actual strontium-90 variation in adult bone is less than that obtained from the short-term experiments but is still far from uniformity. The uncertainty on these factors (Table 3) can be reduced in the future as samples of higher activity level become available.

DISCUSSION

(a) *General pattern*

The data show that: (1) the distribution in adults is markedly nonuniform, (2) the distribution in the fetuses and newborns appears to be uniform, and (3) the overall concentration is much higher in children than in adults. These results reflect the fact that the growing bones of young children are metabolically active throughout, whereas the bones of the adult skeleton have lower and more variable activity. From the dietary concentration of strontium-90, it is possible to compute that the equilibrium bone

Table 1. *Strontium-90 in adult skeletons* 1956–1957, *New York City* (pc Sr^{90}/g Ca)

Vertebra	Rib	Long bone average*	Total skeleton
0.067 ± 0.008	0.05 ± 0.007	0.02 ± 0.003	0.064 ± 0.009
0.18 ± 0.02	0.13 ± 0.01	0.03 ± 0.007	0.163 ± 0.010
	0.10 ± 0.01		
0.14 ± 0.02	0.10 ± 0.02	0.03 ± 0.003	0.052 ± 0.004
	0.04 ± 0.02		
0.05 ± 0.02	0.02 ± 0.02	0.04 ± 0.015	0.038 ± 0.006
0.16 ± 0.03	0.04 ± 0.03	0.04 ± 0.005	0.047 ± 0.006
0.07 ± 0.02	0.03 ± 0.01	0.02 ± 0.003	0.021 ± 0.004
0.03 ± 0.01	0.02 ± 0.02	0.03 ± 0.003	0.014 ± 0.005
0.12 ± 0.01	0.08 ± 0.01	0.05 ± 0.01	0.028 ± 0.003
0.10 ± 0.01	0.03 ± 0.02	0.02 ± 0.005	0.024 ± 0.005
0.17 ± 0.03	0.12 ± 0.03	0.06 ± 0.01	0.042 ± 0.005
0.07 ± 0.01	0.14 ± 0.02	0.01 ± 0.01	0.039 ± 0.005
0.13 ± 0.02	0.05 ± 0.02	0.08 ± 0.05	0.06 ± 0.02
0.09 ± 0.01	≤0.03	0.04 ± 0.007	0.096 ± 0.007
	0.03 ± 0.02		
0.06 ± 0.02	0.03 ± 0.03	0.02 ± 0.003	0.107 ± 0.008
	≤0.02		
≤0.02	0.03 ± 0.02	0.02 ± 0.005	0.072 ± 0.006
	0.03 ± 0.03		
0.03 ± 0.02	0.02 ± 0.01	0.02 ± 0.005	0.126 ± 0.008
0.02 ± 0.03	0.06 ± 0.01	0.05 ± 0.01	0.007 ± 0.012
	0.04 ± 0.02		
0.12 ± 0.01	0.17 ± 0.02	0.02 ± 0.007	0.071 ± 0.006
	0.10 ± 0.01		
0.08 ± 0.02	0.06 ± 0.02	0.02 ± 0.003	0.079 ± 0.006
	0.03 ± 0.02		
0.14 ± 0.02	0.04 ± 0.02	0.03 ± 0.003	0.052 ± 0.004
	0.10 ± 0.02		
0.09 ± 0.01	≤0.02	0.03 ± 0.007	0.052 ± 0.007
	0.07 ± 0.01		
0.14 ± 0.01	0.09 ± 0.01	0.03 ± 0.015	0.088 ± 0.007
	0.10 ± 0.01		
≤0.04	0.045 ± 0.020	0.02 ± 0.003	0.062 ± 0.005
	0.101 ± 0.014		
0.112 ± 0.018	≤0.02	0.03 ± 0.01	0.016 ± 0.005
0.068 ± 0.011	≤0.02	0.02 ± 0.005	0.090 ± 0.006
	≤0.04		
0.12 ± 0.02	0.04 ± 0.02	0.02 ± 0.003	0.051 ± 0.005
	0.02 ± 0.03		
0.18 ± 0.01‡	0.048 ± 0.005	0.012 ± 0.003	≤0.07
	0.080 ± 0.007		
0.139 ± 0.020‡	0.114 ± 0.005	0.025 ± 0.005	≤0.06†
0.095 ± 0.004‡	0.095 ± 0.006	0.033 ± 0.011	≤0.06†
Ave. 0.11	0.07	0.03	0.06

* Errors on vertebra, rib and total skeleton are counting errors. In long bone (in which three or more analyses were done) standard error of the mean is given.

† Normalized to early 1957 average total skeleton.

‡ These are composites of five samples for all bones.

Table 2. Strontium-90 concentrations in various bones of the skeletons of children
(pc Sr^{90}/g Ca)

	Vertebra	Rib	Long bone
Newborn	0.49 ± 0.07	0.54 ± 0.16	0.33 ± 0.04
	0.33 ± 0.10	0.07 ± 0.13	0.17 ± 0.08
	0.35 ± 0.08	0.68 ± 0.15	0.34 ± 0.04
	0.50 ± 0.09	0.59 ± 0.17	0.81 ± 0.07
	0.68 ± 0.06	0.97 ± 0.08	0.80 ± 0.04
	0.47 ± 0.06	0.57 ± 0.08	0.78 ± 0.04
	0.28 ± 0.15	0.85 ± 0.21	0.29 ± 0.08
	0.69 ± 0.05	0.44 ± 0.05	0.77 ± 0.06
	1.02 ± 0.07	0.71 ± 0.08	0.81 ± 0.06
	0.34 ± 0.05	0.61 ± 0.05	0.34 ± 0.04
Average	0.52	0.60	0.54
2 months–6 years	0.93 ± 0.40	0.17 ± 0.25	0.89 ± 0.12
	0.65 ± 0.15		0.41 ± 0.08
	0.55 ± 0.09	0.52 ± 0.08	0.65 ± 0.11
	0.75 ± 0.36	0.98 ± 0.24	0.65 ± 0.11
	1.22 ± 0.23	1.55 ± 0.13	0.68 ± 0.08
	0.49 ± 0.30	0.22 ± 0.13	≤0.12
	0.75 ± 0.36	0.98 ± 0.24	
		0.41 ± 0.04	0.24 ± 0.08
	1.22 ± 0.31	1.48 ± 0.14	0.78 ± 0.09
Average	0.94	1.04	0.58
13 year old	0.21 ± 0.03	0.22 ± 0.02	0.13 ± 0.02
	0.16 ± 0.03		0.16 ± 0.02
	0.12 ± 0.05		≤0.05
			0.15 ± 0.03
			≤0.09
			0.19 ± 0.03
			0.12 ± 0.009
			0.46 ± 0.02
			0.08 ± 0.04
			0.30 ± 0.05
			0.07 ± 0.08
			0.11 ± 0.05
Average	0.16	0.22	0.13

Table 3. Relative distribution of strontium-90 in the skeleton at different ages

	Vertebra	Rib	Long bone	Body average
Adult (ave. 32)	1.8 ± 0.1	1.1 ± 0.1	0.5 ± 0.1	1.0 ± 0.1
13 year old	0.7 ± 0.1	1.0 ± 0.1	0.6 ± 0.1	0.8
2 months–6 years (ave. 14)	0.9 ± 0.1	1.0 ± 0.1	0.6 ± 0.1	
Newborn (ave. 10)	0.9 ± 0.1	1.0 ± 0.1	0.9 ± 0.2	

The indicated uncertainties are the standard errors on the mean values after the bones of each individual or integrated set of individuals are normalized. Values are normalized to whole body ash = 1.0 for the adults, and to rib = 1.0 for all others.

Table 4. Ratio of Sr^{90} concentration in particular bone to skeletal average

	Present	Previous estimate[3]	Sr^{85} [4]
Vertebra/ whole skeleton	1.8	3.4	4.8
Rib/ whole skeleton	1.1	1.5	1.5
Long bone/ whole skeleton	0.5	0.8	0.8

level in this period should be between 1 and 1.5 S.U. (micromicrocuries strontium-90 per gram calcium).[3] This is to be compared with the measured levels in the bones of the forty-two individuals reported here, i.e. adult skeletal average 0.06, adult vertebrae 0.11, and young children 0.8. Thus children are somewhat close to being in equilibrium with their diet while even the most active adult bones are far from equilibrium. The values in young children suggest near uniformity with the long bone being slightly lower than the skeletal average. The concentration in the fetal skeleton is significantly less than that in young children in agreement with the fetal protective factor observed in experimental animals by COMAR.[9]

(b) *Change in skeletal distribution in time*

The nonuniformity within adult skeletons would be only a temporal effect on a constant strontium-90 diet, since common strontium is uniformly distributed in the adult skeleton.[5] The present data on strontium-90 are as yet insufficient to measure this trend toward equilibrium. This is due to the fact that (a) total skeletons from the early years are limited in number, and (b) values of strontium-90 concentrations, particularly on long-bone shaft, are too low to measure with adequate precision until 1956. This shift toward equilibrium, however, is definitely seen if the strontium-90 values reported here are compared with those obtained in the short-term strontium-85 experiments of SCHULERT *et al.*,[4] as the data in Table 4 clearly indicate.

(c) *Intra-bone variations*

Considerable intra-bone variations in strontium-90 concentrations are also apparent. Insufficient strontium-90 is present to make radioautographic investigations of discrete areas. However, data from bone segments analyzed indicate, as expected, that the epiphyses, joint areas, and bone extremities generally have higher concentrations than the midsections of the shafts. In the two most recent composite analyses shown in Fig. 1, the joint areas average 57 per cent higher than limb shafts; also rib ends average about 25 per cent higher than the midsections although the data are somewhat scattered. In both of these cases the mid-60 per cent of the bone is taken as the shaft while the rest is included with the joint.

Fig. 2 shows the strontium-90 concentration

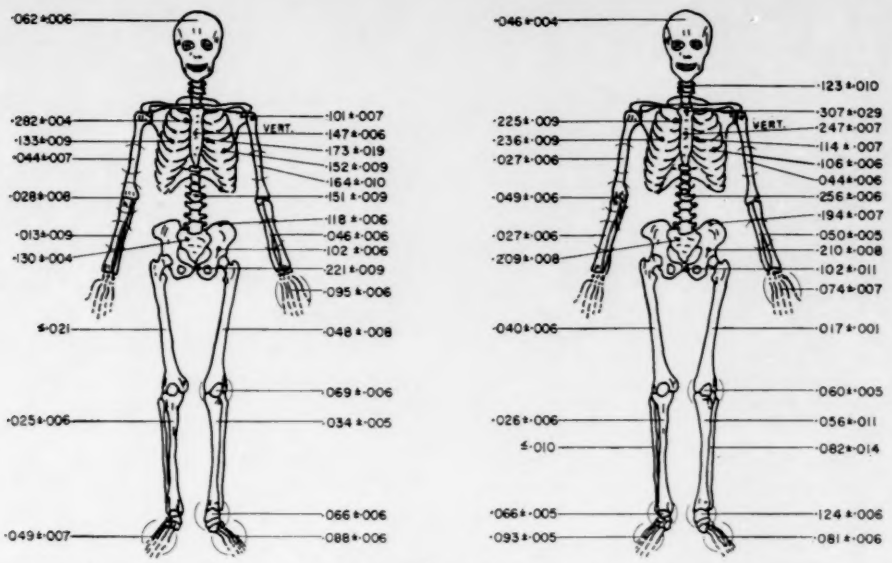

FIG. 1. Sr^{90} variation within skeleton (pc* Sr^{90}/g Ca). Each skeleton represents a composite of five which were analyzed together.

throughout 4–5 cm intervals of tibia and femur from a 13 year old. There appears to be a progressive change from end to midpoint with extreme values differing by about a factor of 4. Bone extremities are generally not analyzed under the Lamont world-wide strontium-90 monitoring program. In previous calculations, insufficient consideration was given to these higher areas of the skeleton in extrapolating from single bone to total body average concentration. Thus it is noted from the summary in Table 4 that while the relative values of the three bone groups are reasonably consistent with the previous data, the value for body average is higher in relation to the other bones than was previously supposed.

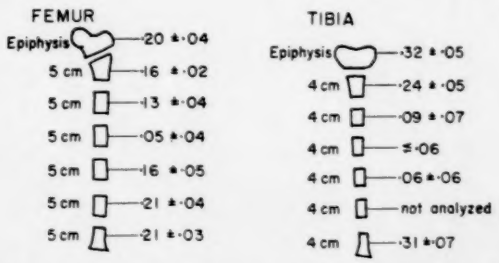

FIG. 2. Intra-bone Sr^{90} variation (pc Sr^{90}/g Ca)

* pc is used throughout to indicate micro microcurie ($\mu\mu c$).

(d) *Radiological implications*

The distribution data on the adult skeleton now make it possible to determine total skeletal load no matter what bone of the body is analyzed. The question arises as to whether the average skeletal concentration or the highest bone concentration is the most important quantity. The investigations of BRUES[10] and FINKEL[11] suggest a threshold does exist for somatic damage. If a threshold exists for the deleterious effects of strontium-90 then the highest concentration in a local area of the skeleton is of primary importance, since this is the area which will first exceed the threshold. However, if there is no threshold and the probability of damage throughout the skeleton is strictly proportional to the radiation received, then the total radiation delivered to the skeleton (or the average Sr^{90}/g calcium) is the important consideration.

Even with a nonthreshold concept, distribution is a consideration in evaluating hazard

since certain skeletal areas are more susceptible to radiation than others. For example, since some leukemia is caused by irradiation of the bone marrow, and since about 40 per cent and 25 per cent of the skeletal marrow is located in the vertebrae and ribs, respectively, it is calculated from the adult distribution data of Table 3 that the marrow is getting twice the radiation as the average received by the whole skeleton and thus this damage is twice as great as the average values indicate even with the linear, nonthreshold concept. Nevertheless, the nonuniform distribution in the adult skeleton is of little consequence in establishing tolerable fallout contamination since the strontium-90 concentration, as noted, is highest in children which have essentially uniform distribution.

It is generally agreed that the elimination of strontium-90 from the body cannot be defined in terms of a simple biological half-life, since the element (in common with the other alkaline earths) is metabolically treated in such diverse manner in different areas of the skeleton.[12] The disappearance of strontium from the skeleton may be considered as taking place from a series of loci, each with its own mean residence time. The distribution data obtained in this investigation show that the residence time for strontium-90 in vertebrae is shorter than that for rib, which in turn is shorter than long-bone shaft (since the metabolically more active bone which accepts the isotope most readily would also lose it most rapidly). However, there are, of course, subcategories of relatively active and inactive areas within each bone type.

SUMMARY AND CONCLUSIONS

(1) Various bones of the skeleton and an aliquot of the total skeletal ash were analyzed for strontium-90 in the case of many adult cadavers from the New York City area. A number of fetuses were analyzed in the same manner as well as individual bones (usually 2–4 per skeleton) from children in the 0–14 age group.

(2) The distribution of strontium-90 is nonuniform in adults with the concentration in vertebrae, rib and long-bone shaft being respectively, 1.8, 1.1 and 0.5 times the body average.

(3) Young children exhibit essentially uniform distribution.

(4) The data permit the determination of total skeletal strontium-90 burden in a population.

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THE FLUX AND SPECTRUM OF COSMIC-RAY PRODUCED NEUTRONS AS A FUNCTION OF ALTITUDE

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(*Received* 13 *January* 1959)

Abstract—A series of measurements of neutron flux as a function of altitude has been made with the following neutron detectors: bismuth fission ionization chamber, proton-recoil proportional counter, and moderated and bare BF_3 proportional counters. In addition, data were also taken with a Simpson pile. Altitudes ranged from sea-level to 40,000 ft and latitudes from 28° to 49°N. Appropriate treatment of the data can be made to yield information about the neutron spectrum and the average neutron energy, and consequently about the dose rate.

I. INTRODUCTION

WHEN one is charged with the responsibility for the assessment of radiation fields present near an intense but shielded source of fast and high-energy neutrons, it is desirable to have a variety of neutron detectors whose sensitivity and efficiency are well known. Much of the effort of the Health Physics Department at the University of California Lawrence Radiation Laboratory is directed toward the development of such detectors.[1,2,3] The construction of a large Bi fission ionization chamber made possible the direct measurement of very small high-energy neutron fluxes,[4] and led to initiation of the experiment to be described.

Although the experimental results have consequences in shielding theory and cosmic-ray physics, these aspects are not discussed here. Instead, our concern is with the results of primary interest to a health physicist—the determination of the neutron dose rate.

II. EXPERIMENT

When primary cosmic rays enter the earth's atmosphere and interact with it, a variety of particles—including neutrons—are produced. (It is unlikely that many neutrons are present in the primary cosmic rays because of their 12 min half-life and also because of the lack of suitable accelerating mechanisms.) The attenuation of high-energy cosmic-ray produced neutrons in the atmosphere is analogous to the attenuation of accelerator- or pile-produced neutrons in concrete. That this analogy holds is because after the transition region is passed the attenuation of neutrons of all energies is controlled by the attenuation of the most penetrating primary particles and secondary neutrons. In addition, for neutrons whose energy is more than a few MeV, concrete and air exhibit nearly the same attenuation property, on the basis of equal area density (g/cm^2), because they both consist of materials of low atomic number. Thus, after equilibrium is attained, one should expect a continuous neutron spectrum, the shape of which does not vary with thickness, and the total intensity of which decreases with an attenuation rate characteristic of the most penetrating neutrons. Moreover, the spectrum should be approximately a simple reciprocal power of neutron energy, near E^{-1}, as would be inferred from slowing-down concepts.[7]

It is possible to determine such a continuous neutron spectrum in air or concrete with fair precision by measuring the absolute flux at definite energy intervals within the spectrum. Accordingly we calibrated a BF_3 counter, a polyethylene-lined proton-recoil counter, and a Bi fission ionization chamber in known neutron fluxes. The data are given in Table 1.

These counters and a Simpson pile[5] (for intercomparison with other experiments) were

Table 1. *Calibration of detectors for determining a neutron spectrum in air or concrete*

Detector	Energy interval of effective response (MeV)	Response in units of
BF_3 counter (bare)	10^{-9} and up	counts per neutron/cm^2 (varies as $1/V$)
BF_3 counter + 5 cm paraffin covered by 0.020 in. Cd jacket	0.03 to 14	counts per neutron/cm^2
CH_2-lined proportional counter	0.03 to 30	counts per MeV/cm^2
Bi fission ionization chamber	100 to 1000	counts per neutron/cm^2

taken to the 10,000 ft station at the University of California High-Altitude Research Facility in the White Mountain Range in south-eastern California in the summer of 1956. Subsequently they were flown in a B-36 airplane operating from Kirtland Air Force Base, Albuquerque, New Mexico, at altitudes ranging from 5000 to 40,000 ft. Figure 1 shows a curve of counting

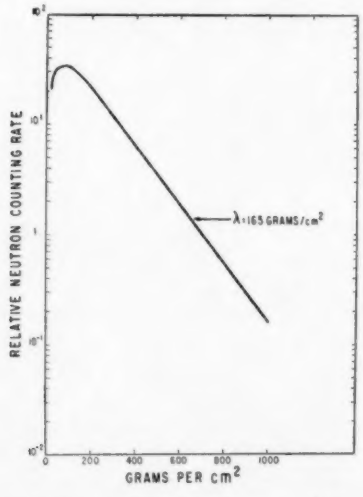

FIG. 1. Curve of relative neutron counting rate vs. depth in the atmosphere, showing the slope which is typical of all our detectors

rate vs. atmospheric depth that is typical of all our detectors. The slope of about 165 g/cm^2 for a $1/e$ reduction in intensity is also typical of the attenuation in concrete of the 270-MeV neutrons produced by the 184 in. synchrocyclotron and of neutrons in the BeV energy range produced by the Bevatron. Good agreement was obtained between the counting rate at 10,000 ft on the ground and in the airplane, tending to show that the effect of ground albedo was small. In this experiment discrimination against counts caused by charged particles in the polyethylene-lined counter and the Bi fission chamber was provided by the output of a blanket of Geiger tubes used in anticoincidence.

Fig. 2 shows the neutron spectrum obtained at 10,000 ft. That the shape of this spectrum is independent of altitude is evidenced by the fact that all the neutron detectors, regardless of energy sensitivity, gave the same slope for curves similar to Fig. 1. This neutron energy spectrum $\phi(E)$, is obtained from the counting rates of the various detectors in the following way. The counting rate C_x of detector x is given by

$$C_x = \int \varepsilon_x(E)\phi(E)\, dE$$

where $\varepsilon_x(E)$ is the efficiency of detector x in counts/neutron per cm^2. The absolute efficiencies of all of the detectors have been measured over a wide range of energies by using several calibrated neutron sources. The neutron energy spectrum is determined by a trial and error solution to the above integral. The intensity of $\phi(E)$ is changed in different energy regions until the calculated counting rates of all detectors agree with the experiment. Below 10 KeV the shape of the spectrum can be calculated by neutron slowing down and capture theory. The peak in the spectrum at about 1 MeV is due to the production of evaporation neutrons.

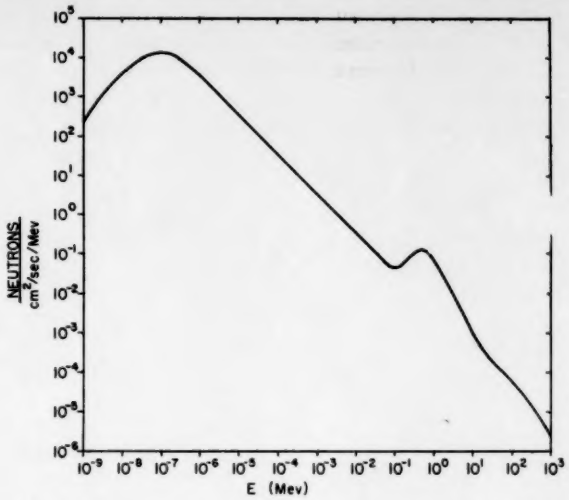

FIG. 2. Neutron energy spectrum from cosmic rays at 10,000 ft.

III. DOSE CALCULATIONS

The dose rate due to neutrons at 10,000 ft is given by

$$\text{dose rate} = \int_{10^{-9}\text{ MeV}}^{10^{3}\text{ MeV}} n(E)1/A(E)\,dE$$

where $n(E)$ is the neutron flux at energy E and $A(E)$ is the flux at energy E per dose-rate unit. This integral may be approximated by

$$\text{dose rate} \approx \Sigma\, \overline{N_i(E)}\; \overline{[1/A_i(E)]}\; \Delta E$$

Values for $[1/A_i(E)]$ can be developed from data found in NBS Handbook 63.

Fig. 3 shows the percentage of the total neutron dose rate at 10,000 ft vs. ΔE for the spectrum given in Fig. 2. The sum, in units of rem/year, is 0.195 The rem dose per year at any "pressure altitude" past the transition region can be calculated by referring to Fig. 1, which indicates that the neutron spectrum is constant and thereby demonstrates the altitude dependence of dose rate. Table 2 gives neutron

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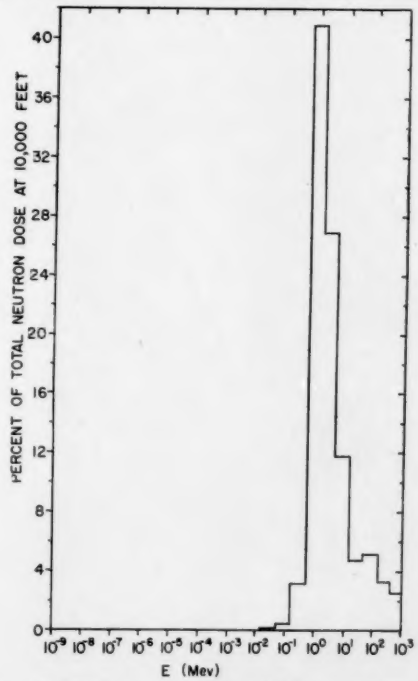

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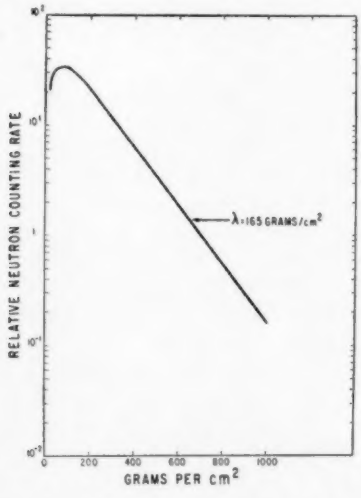

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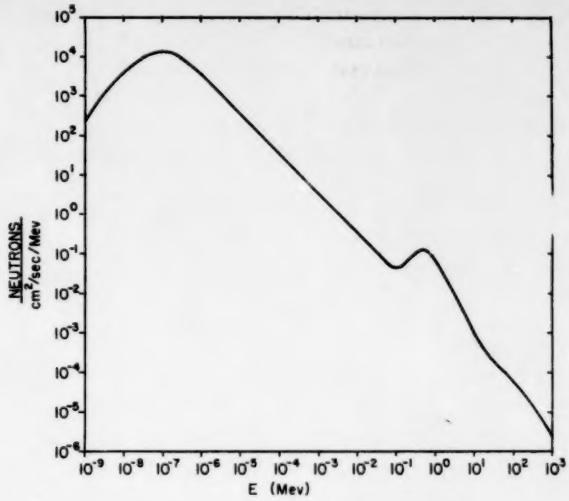

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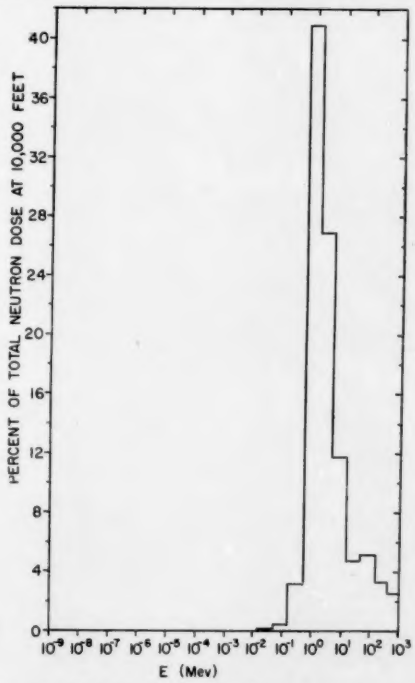

FIG. 3. Histogram giving the percent of total neutron dose vs. ΔE at 10,000 ft.

dose rates at the pressure altitudes of interest.

TOBIAS gives dose rates that are about one-half these values.[6] However, his estimate is based on the counting rate of a balloon-supported BF_3 counter, and he had no direct measurement of fast-neutron flux.

Also Fig. 3 shows that 75 per cent or more of the neutron dose rate in an equilibrium spectrum in air or concrete may be determined by using a polyethylene-lined proportional counter and bare and moderated BF_3 counters.

IV. CONCLUSIONS

Conclusions of interest to health physicists are as follows:

(1) Although the shapes of the neutron spectra in air and in concrete may be significantly different at energies below 10^{-2} MeV, this does not strongly affect the neutron dose rate. In any event, the determination of the thermal-neutron dose rate is not difficult.

(2) By far the largest contribution to total neutron dose comes from neutrons in the energy interval from 0.10 to 30 MeV. Furthermore, this portion of the neutron dose can be determined easily. Refer to Table 1 and notice that the energy regions of response of the polyethylene-lined counter and of the BF_3 counter with 5 cm of paraffin covered by 20 mil Cd are essentially the same. Also notice that the polyethylene-lined counter responds in units of counts per MeV/cm^2, while the moderated BF_3 counter responds in units of counts per neutron/cm^2. Therefore if measurements coincident in time and location are made with these two detectors, one may immediately determine the neutron flux and the mean energy per neutron,

$$\text{MeV/cm}^2 \text{ per count} \div (\text{neutrons/cm}^2 \text{ per count}) = \text{MeV/neutron},$$

in the energy interval of greatest importance for health physics purposes.

Acknowledgment—This work was done under the auspices of the U.S. Atomic Energy Commission.

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CALCULATIONS OF ENERGY LOSS UNDER THE BIAS IN FAST NEUTRON DOSIMETRY

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(*Received* 31 *December* 1958)

Abstract—The amount of energy loss under the bias in an integrating fast neutron dosimeter is treated by considering an idealized model of a dosimeter which has been considered previously by PETROV. It is shown that when one includes that portion of the energy deposited in the chamber which is recorded by an integrating circuit, one finds much smaller losses than are found by PETROV.

THE quantity of energy loss under the bias in an integrating fast neutron dosimeter has been considered by WAGNER and HURST.[1,2] They find experimentally that the fractional energy loss under the bias is always much less than unity for bias values necessary in practical cases; usually it is only a few per cent. PETROV,[3] on the other hand, considers losses in an idealized parallel plate dosimeter and finds that they may be much larger than the experimentally determined values. It is the purpose of this paper to reconsider the problem treated by PETROV and to show that his model in reality yields smaller losses below the bias than the values quoted by him. As suggested in the preceding paper,[2] this is apparently due to a misconception on his part as to the exact function of the binary integrator.

The model considered by PETROV is that of neutrons normally incident on a thick plane parallel radiator of thickness greater than the range of the most energetic protons generated therein. Protons are ejected from the radiator into a narrow gas-filled gap (see Fig. 1). Both radiator and gap are considered to be effectively infinite in lateral extension. One wishes to calculate the total energy deposited in such a chamber by protons generated in the walls and gas and expending energy in the gas at least as great as the bias energy. Clearly, a realistic chamber of finite lateral extent will have

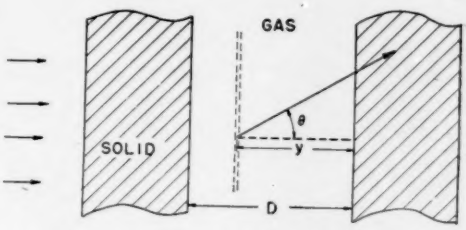

FIG. 1. Schematic representation of a parallel plate fast neutron dosimeter.

generated in it a distribution of proton energies which will be more heavily populated in the low-energy region than in the case of an infinite chamber. Hence the bias losses calculated for this idealized model should always be less than one would calculate for a real chamber. The following notation is employed;

D = gap spacing
A = area of radiator normal to the neutron beam
N_g = number of hydrogen atoms per cm^3 of gas
N_R = number of hydrogen atoms per cm^3 in radiator
ϕ = incident neutron flux
E = energy of incident neutrons
$\sigma(E)$ = n–p scattering cross-section at energy E
$E_p(\theta)$ = energy of protons recoiling at the angle θ relative to the beam direction

* Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

E_e = energy lost in gap by a proton

$\mu = \cos\theta$

y = distance from gap wall at which proton recoil may occur.

A range energy relation of the form $R = kE^n$ will be assumed for protons in both gas and radiator. Following HURST *et al.*[4] and PETROV,[3] n will be taken to be 1.72–1.73. Clearly, if $N_g k_g = N_R k_R$, where k_g and k_R refer to gas and radiator respectively, one may neglect the difference in density between radiator and gas in this simple one-dimensional case. Since this

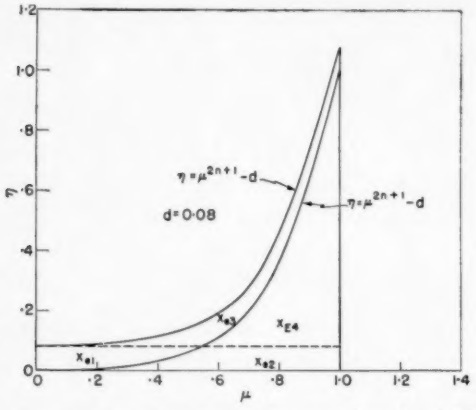

FIG. 2. Plot of regions in the η–μ plane in which different expressions for the energy loss must be employed.

is true to a good approximation in the gas-wall combinations considered in references 3 and 4, and in any chamber designed in accordance with the Bragg–Gray principle, the equivalent problem of the energy deposited between parallel planes a distance D apart in an infinite medium of gas is considered.

Consider protons recoiling at the angle θ from an n–p collision occurring at a distance y from the far wall of the gap (Fig. 1). Since

$$E_p(\theta) = E\mu^2 \qquad (\mu \geq 0)$$

and the fraction of recoils occurring at y in the interval $d\mu$ at μ is $2\mu\, d\mu$, the number of protons liberated in dy at y and $d\mu$ at μ which lose the energy E_e in the gap is

$$d^2N = \phi N_g \sigma A 2\mu\, d\mu\, dy \qquad (1)$$

Equation (1) may be rewritten conveniently in terms of the dimensionless variables, $\eta = y/kE^n$, and $\xi = 2\phi N_g \sigma A k_g E^n$. One then has

$$d^2N = \xi\mu\, d\mu\, d\eta \qquad (1a)$$

The energy $E_e = x_e E$ deposited in the gap by a given proton at μ and η may be written in terms of η and μ from the range-energy relation. For example, if $y \leq D$, the recoil may or may not strike the wall. If it does not, the energy loss E_e is just

$$E_e = E\mu^2$$

and the range of θ for which this condition obtains may be found by referring to Fig. 1 and is given by

$$\theta_0 \leq \theta \leq \pi/2$$

where

$$\cos\theta_0 = \mu_0 = \frac{y}{kE_p{}^n(\theta_0)} = \frac{\eta}{\mu_0{}^{2n}}$$

Then one finds for x_e in this range of μ

$$x_{e1} = \mu^2 \qquad d \geq \eta \geq \mu^{2n+1} \qquad (2a)$$

where $d = D/kE^n$.

If the recoil originates in the gas ($\eta \leq d$) and strikes the wall, the energy loss in the gas is the total energy of the recoil $E\mu^2$, minus the energy deposited in the wall, $[kE^n\mu^{2n} - (y/\mu)]^{1/n}$. Then in terms of the dimensionless variables

$$x_{e2} = \mu^2 - \left(\mu^{2n} - \frac{\eta}{\mu}\right)^{1/n}$$

$$\eta \leq \min\,(d, \mu^{2n+1}) \qquad (2b)$$

where min (a, b) and max (a, b) indicate that the smaller and larger values, respectively, of a and b are to be used. Proceeding in a similar way one finds the following expressions for x for recoils which originate in the near wall and do not enter [equation (2c)] or do enter [equation (2d)] the far wall.

$$x_{e3} = [\mu^{2n} - (\eta - d)/\mu]^{1/n}$$

$$\max\,(d, \mu^{2n+1}) \leq \eta \leq \mu^{2n+1} - d \qquad (2c)$$

$$x_{e4} = [\mu^{2n} - (\eta - d)/\mu]^{1/n} - (\mu^{2n} - \eta/\mu)^{1/n}$$

$$d \leq \eta \leq \mu^{2n+1} \qquad (2d)$$

Fig. 2 shows the boundaries of the four regions in which equations (2a)–(2d) are valid, for the case $d = 0.08$. The total energy deposited

in the gap by protons losing energy at least as great as $B = xE$ in the gap, is given by the energy E_e ($E_e \leq B$) times the number of protons $\xi\mu\, d\mu\, d\eta$ losing this energy summed over all permissible values of η and μ. Thus one finds

$$E_T = E\xi\left[\int_{\sqrt{x}}^{d^{1/2n+1}} \mu\, d\mu \int_{\mu^{2n+1}}^{d} d\eta\, x_{e1} + \int_{\sqrt{x}}^{d^{1/2n+1}} \mu\, d\mu \int_{\mu^{2n+1}-\mu(\mu^2-x)^n}^{\mu^{2n+1}} d\eta\, x_{e2} + \int_{d^{1/2n+1}}^{1} \mu\, d\mu \int_{\mu^{2n+1}-\mu(\mu^2-x)^n}^{d} d\eta\, x_{e2} + \int_{\sqrt{x}}^{d^{1/2n+1}} \mu\, d\mu \int_{d}^{\mu^{2n+1}-\mu x^n+d} d\eta\, x_{e3} + \int_{d^{1/2n+1}}^{1} \mu\, d\mu \int_{\mu^{2n+1}}^{\mu^{2n+1}-\mu x+d} d\eta\, x_{e3} + \int_{d^{1/2n+1}}^{1} \mu\, d\mu \int_{d}^{\mu^{2n+1}} d\eta\, x_{e4}\right] \quad (3)$$

Note that this expression for E_T is valid only as long as $x \leq 1 - (1-d)^{1/n}$, i.e. as long as no recoil originating in the near wall and entering the far wall loses less than the bias energy B in the gas. When x is larger than this value, the expression for E_T involves the solution of a transcendental equation in the limits of integration and cannot be given in analytical form. We will not consider this case further here.

Equation (3) may be reduced to the following explicit form

$$E_T = \xi E\left[\frac{d}{4}(1-x^2) - \frac{1}{(n+1)(2n+5)} \times (1-x^{n+5/2}) - \frac{n}{3(n+1)}\, x^{n+1}(1-x^{3/2}) + \int_{\sqrt{x}}^{1} \mu^4(\mu^2-x)^n\, d\mu - \frac{n}{n+1} \times \int_{\sqrt{x}}^{1} \mu^2(\mu^2-x)^{n+1}\, d\mu\right] \quad (4)$$

Note that E_T reduces to the value $\xi E d/4$ when $x \to 0$, as it must. Using the definition of WAGNER and HURST,[2] we set $f(x) = 1 - (4E_T x)/(\xi E d)$ to be the fractional part of the total energy loss below the bias energy $B = xE$. Expressing the integral terms in ascending series in powers of x, one has

$$f(x) = x^2 + \frac{4}{d}\, g(x) \quad (5)$$

$$x < 1 - (1-d)^{1/n}$$

where

$$g(x) = \frac{n}{3(n+1)}\, x^{n+1} + \left[\frac{2n+3}{3} \times \sum_{r=0}^{\infty} \frac{(-1)^r(n)(n+1)\ldots(n-r+1)}{r!(2n+5-2r)} - \frac{1}{(2n+5)(n+1)} - \frac{n}{3(n+1)}\right] x^{n+5/2} + \sum_{r=2}^{\infty} \frac{r-1}{r!}\, \frac{n(n-1)\ldots(n+2-r)}{(2n+5-2r)}\, (-x)^r \quad (6)$$

One may show that equation (5) is also valid in the case $d > 1$. For the case under consideration where $n = 1.73$,

$$g(x) = 0.211233x^{2.73} - 0.153314x^{4.23} + 0.193946x^2 - 0.171125x^3 - 0.092658x^4 + 0.009373x^5 + 0.001928x^6 + 0.000691x^7 + 0.000316x^8 + \ldots \quad (7)$$

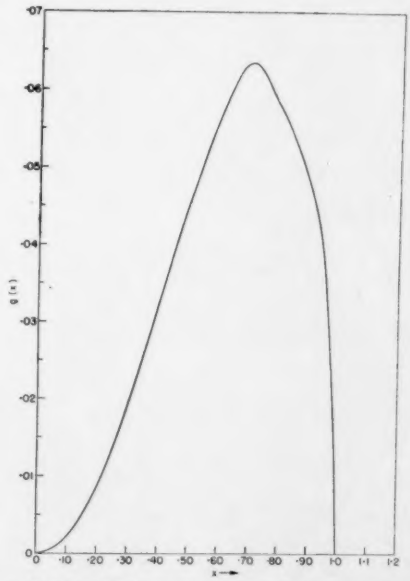

FIG. 3. Plot of a function occurring in the expression for energy loss under the bias, equation (5).

Table 1. Comparisons of losses under the bias in the idealized dosimeter

D (cm)	E (MeV)	B (MeV)	d	x	$(1-f)$ Petrov	$(1-f)$ Eq. (5)
1	1	0.05	0.524	0.05	0.884	0.993
	1	0.10	0.524	0.10	0.767	0.974
1	2	0.05	0.158	0.025	0.875	0.997
	2	0.10	0.158	0.05	0.748	0.984
1	3	0.05	0.075	0.0167	0.845	0.998
	3	0.10	0.075	0.0333	0.691	0.988
4	2	0.10	0.633	0.05	0.890	0.994
	2	0.20	0.633	0.10	0.780	0.977
4	5	0.10	0.129	0.02	0.885	0.997
	5	0.20	0.129	0.04	0.770	0.988

Fig. 3 shows a plot of $g(x)$. The values of $1-f$ given in Table 1 were calculated from equation (5). Also shown are values given by PETROV for this quantity. The constant k_g has been taken the same as that used by PETROV for the purposes of comparison, namely 1.91 cm $(\text{MeV})^{-1.73}$, and is appropriate to acetylene at 15°C and 760 mm Hg.[3] Apparently, as suggested by WAGNER and HURST,[2] PETROV has assumed that the fast neutron integrator pulse misses not only those pulses having energies less than the bias but misses also that part of a given pulse which lies below the bias value.

Acknowledgment—The author would like to express his thanks to G. S. HURST, Oak Ridge National Laboratory, for suggesting this problem.

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NOTES

Integral-Phospor Counting of α-Emitters*

(*Received* 16 *February* 1959)

SILVER-ACTIVATED zinc sulfide powder, a standard material for the scintillation detection of α-activity, has been reported to remove polonium and the decay products of radon from solution,[1] and has been found to remove plutonium and uranium from solution. The zinc sulfide powder, which settles out of solution, is a radioactive phosphor, the scintillations from which may be detected with a bare multiplier phototube and counted by the usual methods. Additional amplification of the electrical signal is unnecessary. This process, if quantitative, might prove to be a useful method for the assay of α-emitters. This paper presents results of a preliminary study to determine the possibilities of such use.

The efficiency of removal of plutonium by zinc sulfide is pH-dependent, as shown by the data in Table 1; 20,000 d/min of plutonium-239 in 1 ml of 0.5 N HCl were added to each of five flat bottom, clear glass vials. The pH of each solution was adjusted, by adding base, to provide a series ranging from pH 4 to pH 12. Total volumes in these vials ranged from 2 to 10 ml. Silver-activated zinc sulfide (400 mg) was added to each vial, the vials were shaken briefly, and the powder in each vial was permitted to settle for 2 hr. The vials were then set on the face of an upright multiplier phototube and the scintillations counted.

From the 7 ml of solution in the pH 4 vial, 1 ml was removed, dried on a foil and counted. Corrected for background and counting efficiency, this sample indicated that the solution retained about 3.6 per cent of the initial activity. The plutonium activity added to the vial may have been in error by as much as 5 per cent, so the removal might be complete.

Included in Table 1 are data from a similar experiment using polonium-210, in which the settling time for the powder was 15 hr following the addition of 70 mg of ZnS(Ag). The overall detecting efficiency is less than that for plutonium because of the lesser amount of zinc sulfide, as is described below. Half the total solution in each vial was analyzed for polonium by standard plating and counting procedures.

These data indicate that at pH 4, both plutonium and polonium are removed to the same extent, although the amounts of zinc sulfide were quite different. Removal is greatest at low and high pH values, where more than 95 per cent of the material is removed. An adsorption mechanism is indicated, in which case the extent of removal is probably independent of the element used. It is probable that less than 70 mg of ZnS(Ag) is required for quantitative removal, but it is shown below that more than this amount is required for optimum counting efficiency. (The amounts of plutonium, 0.15 μg, and polonium, 2 pg, used are considerably less than that of zinc sulfide. However, since the bulk of the powder remains insoluble, surface area ratios may be important; the surface area of the powder has not been determined.)

Zinc sulfide powder is not transparent and absorbs some fraction of the emitted light. However, a thicker layer of powder (up to the range of the α-particle) will produce a brighter scintillation than will a thin layer. Both of these effects act to produce a continuum of pulse amplitudes from the scintillations produced by the monoenergetic α-particles. There is, therefore, an optimum thickness which will produce both a maximum pulse amplitude and a maximum counting rate. An experiment, using polonium at pH 2, in which zinc sulfide was added in increasing amounts, with activity determined between additions, indicated that about 420 mg of zinc sulfide yielded the highest counting efficiency. The vials used had a bottom area of 3.5 cm^2, so that the optimum thickness of the phosphor layer is about 120 mg/cm^2. The curve of thickness vs. count rate (shown in Fig. 1) has a broad maximum so that the amount added is not critical. The maximum overall counting efficiency was found to be 72 per cent for both polonium and plutonium with about 400 mg of ZnS(Ag).

An efficiency greater than 72 per cent might be obtained by providing additional amplification of the signal, since the count rate-high voltage characteristic was still rising at the highest voltage used. (A more recent version of this counter, used for uranium

* This paper is based on work performed under contract with the United States Atomic Energy Commission at the University of Rochester Atomic Energy Project, Rochester, New York.

Table 1. *Removal of plutonium-239 and polonium-210 from solution by zinc sulfide*

pH	Plutonium + 400 mg ZnS(Ag)		Polonium + 70 mg ZnS(Ag)	
	overall counting efficiency (%)	in solution, (% of initial)	overall counting efficiency (%)	in solution, (% of initial)
2			58	1.7
4	72	3.6	53	3.7
6	64		43	34
8	64		47	5.2
10	65		24	37
12	72		55	1.7

analysis, has a greater amplification in the phototube circuit. This counter exhibits a plateau about 200 V wide, with a slope of 3 per cent per 100 V, and a background of a few counts per hour. When operated in the mid-plateau region, uranium is counted with 100 per cent efficiency.)

Drying of the sample in the vial increased the efficiency to about 95 per cent. Centrifugation of the powder was also effective, and provided an efficiency greater than 90 per cent. This increased efficiency must be due to compacting of the phosphor layer

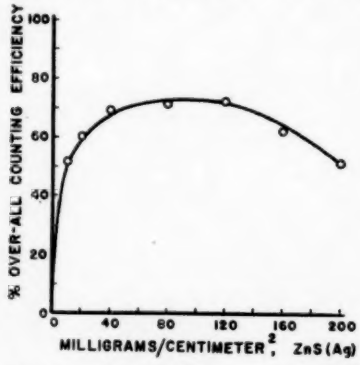

FIG. 1. Counting efficiency vs. Layer thickness.

since the increase in efficiency is greater than could be due to the addition of the non-bound material to the phosphor.

This method of assay appears to hold promise for those situations in which only one isotope is present, or if total α-activity only is desired. Since the energy resolution of this detector is very poor, analysis of mixtures is not feasible. It may be applicable to the biological assay of α-emitters, although some difficulties with interference should be anticipated. It provides a counting method which is simple and direct in application, provides a high counting efficiency with low background, and shares with liquid scintillation counting methods the advantages of internal standardization.

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Acknowledgment—The author would like to acknowledge the assistance of Mrs. PEARL MILLER in the preparation of the samples.

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Some Unusual X-Radiation Dosimetry Problems associated with Radar Installations

(*Received* 19 *January* 1959)

HIGH power radar transmitters, such as those used in missile tracking and early-warning networks, present some rather unusual radiation protection problems. There are two types of electromagnetic radiation generated simultaneously; one is the microwave radiation the transmitter is designed to generate, and the other is X-radiation, in this case a non-functional by-product.

Since microwaves require radio rather than radiological techniques to measure, the health physicist would prefer to leave this field to the radio-frequency engineer. However, it has been found that *r.f.* has considerable effect on electronic circuits, and that ionizing radiation survey equipment

is subject to serious errors when used near operating radar equipment.[1,2]

Briefly, the entire survey instrument, including any external probes or cables should be electrically shielded; using conducting glass for meter windows and conducting gasketing, case materials and finishes, all with very low resistance. Rather than redesign the cases and components of commercial equipment, it was found possible to reduce *r.f.* pickup sufficiently by enclosing critical portions of the unit in a copper wire-mesh container. Where knobs or handles protruded, good electrical contact with the screening was essential.

However, the addition of wire mesh screening around the ionization chamber introduced energy dependence in an otherwise air equivalent chamber wall, especially for low energy X-radiation. Accordingly, tests were made to determine what corrections were necessary.

Eighty-mesh copper screening was fitted around the chamber of a conventional cutie-pie survey instrument with a pliofilm end window. This mesh was found adequate for shielding 3 cm microwave fields of up to 100 mW/cm^2.

The energy of the X-radiation produced in the magnetrons, thyratrons, and klystrons in question ranged from 30 kVp to about 150 kVp. A medical X-ray machine was used to produce X-rays with peak potentials in this range. It was thus possible to note the difference in the instrument reading for the same exposure when the copper screening was removed from the chamber. From these data a curve was plotted as in Fig. 1 showing the required correction factors when using 80-mesh screening.

A most interesting problem in radar X-ray dosimetry stems from the pulsed nature of the source and the often extreme intensity of the radiation during the pulse. Average dose rates as high as 300 r/hr have been measured 1 ft away from one klystron collector operated at a duty cycle of 0.0015. Thus, during a pulse, an ionization chamber would be subjected to about 200,000 r/hr.

The problem was to insure that the potential across the ion chamber was adequate for complete saturation for the highest peak intensity fields in which it would be used. The most direct experimental approach would be to expose a survey instrument in the X-ray field in question where a reading of about 90 per cent of its range was obtained. Then, if no significant increase in reading resulted as the chamber potential was doubled, it would be safe to conclude that the chamber would be saturated over its full range.

Alternatively, it was found possible to predict instrument performance before a pulsed X-ray source was conveniently available. Two approaches were taken, one theoretical and the other experimental.

For the theoretical approach, the two major causes of non-saturation effects were considered, ion recombination and space charge.

Small recombination losses may be calculated for a specific chamber under the following assumptions, which were reasonable in our case.

(1) Ions are produced uniformly throughout the chamber at some rate (N) per unit volume per unit time.

(2) The ion mobilities used are those for negative ions, not electrons; since for most of their life in an air ionization chamber the electrons will be attached to neutral atoms and act as negative ions.

Small fractional losses due to recombination in an air ionization chamber may be approximated by the following expression:[3]

$$f = \frac{N\sigma(\ln y)^2 a^4}{\mu_+\mu_- V^2(y^2-1)}\left[\frac{y^6-1}{24} - \frac{y^2(y^2-1)}{8}\right]$$

where f is the fractional loss
N is the ion production rate per unit volume
σ is the recombination constant
a is the radius of the center electrode
ay is the radius of the outer electrode
μ_+ and μ_- are the ion mobilities
V is the chamber potential.

In a typical case, the instrument was designed such that:

$$a = 0.365 \text{ cm}$$
$$y = 11$$
$$V = 60 \text{ V}$$

For air

$$\sigma = 2 \times 10^{-6} \text{ cm}^3/\text{sec}$$
$$\mu_+ = \mu_- = 2.7 \text{ cm}^2/\text{V-sec}$$

Using these values

$$f = 2.5 \times 10^{-9} N \text{ cm}^3\text{-sec}$$

In air, the value of N for the radiation encountered may be found from the conversion factor

1 r/hr produces about 5.8×10^5 ion pairs/cm^3-sec.

The range of the instrument was 2.5 r/hr, and the shortest duty cycle of concern was 0.05. Thus, it would be expected that a maximum full range indication on the instrument would result for a pulse intensity equivalent to 50 r/hr, and N would be about 2.9×10^7 (ions)/cm^3-sec. The recombination loss was therefore found to be about 13 per cent at the highest reading obtainable on the instrument.

Considering space charge: there is a critical

charge density due to ionization in a given chamber volume at which the potential gradient at the electrode is depressed to zero and the chamber will not be saturated for higher radiation intensities. An expression for the chamber voltage at which this occurs was developed by LICHTENSTEIN[4] as follows:

$$V = \left(\frac{\sigma}{2\varepsilon_0\mu}\right)^{1/2}\left(\frac{a^2+b^2}{2}\right)\ln\left(\frac{a^2+b^2}{2ab}\right)$$

where V is the critical voltage
σ is the current density (A/cm^3)
a is the inner electrode radius
b is the outer electrode radius
ε_0 is the permittivity of free space
μ is the ion mobility

For a typical chamber

$$V = 60 \text{ V}$$
$$a = 0.365 \text{ cm}$$
$$b = 4 \text{ cm}$$

Solving for σ, the critical current density was about 10^{-11} A/cm^3. This would require a pulse intensity equivalent of about 100 r/hr, which was well above the effective intensity range of the instrument considered.

Note, however, that in some installations, where the duty cycle may be 0.001, 2500 r/hr peak would be required to produce an average dose rate of 2.5 r/hr. In this case, a higher chamber voltage may be needed to achieve saturation. A more detailed analysis would be required in such cases particularly with regard to the mechanics of ion recombination with pulsed radiation.

A check on this analysis was made by exposing the chamber to dose rates equivalent to the peak dose-rate required to produce an average of 2.5 r/hr for several different duty cycles. A Beckman micromicroammeter was used to measure ionization current while the chamber potential was changed. A Victoreen R-Meter was used to measure dose rates in a cobalt-60 γ-ray field. Saturation curves were plotted as shown in Fig. 2. With 60 V potential, the loss due to non-saturation was about 14 per cent at 45 r/hr and 45 per cent at 108 r/hr. These data confirmed the theoretical predictions.

Additional tests are planned to determine saturation characteristics at higher intensities with the application of additional chamber voltage; and field tests will be conducted under actual pulsed X-ray exposures for cross calibration with Victoreen R-chambers and film dosimeters. We shall also test pocket ionization chambers for possible non-saturation and r.f. effects.

It is hoped that the results of this work will lead to the design of ionization chamber instruments that the radar technician or engineer can use with the same convenience that the radiographer now enjoys.

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NEWS

News Editor: SAUL J. HARRIS

Health Physics in the News

THE following items concerning Health Physics have appeared in newspapers, public press or technical journals during the last quarter.

Minnesota regulates reactors

On December 4, 1958 the Minnesota State Board of Health adopted Regulations on Ionizing Radiation including the registration of radiation sources, and for the first time by any state, the filing of a reactor hazards report with the state Department of Health. The regulations, which make Minnesota the sixteenth state to require registration of hazardous radiation sources with a state, stipulate that "no reactor shall be operated in this state until the owner has received, and is in the possession of" an approval from the Board of Health. The regulations also ban shoe fluoroscopes.

Additional training courses offered

On May 14–15, 1959, the Training Program of the Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, offered a two-week course in the "Sanitary Engineering Aspects of Nuclear Energy".

On May 4–29, 1959, the Institute of Industrial Medicine at N.Y.U.'s Post Graduate Medical School, New York, N.Y., offered a course in "Radiation Hygiene Measurements."

Contamination incident reported

A case involving the spilling of radioactive material on the hands of a Texas youth in mid-November 1958 received newspaper and national news magazine attention. The youth, JOHNNY PIERCE of Tyler, Texas, reportedly contaminated his hands with radium after opening a medical radium capsule attempting a home experiment. The boy first said he had obtained some cobalt-60 without a license, but further investigation proved the contamination to be radium. While contaminated the boy managed to contaminate about a dozen business places in Tyler.

AEC regulations amendments proposed

In the Federal Register of October 31, the AEC proposed amending its regulations in Part 30, Title 10, C.F.R. to change the basis for exempting manufactured items containing radioactive materials. The new proposal is to exempt products depending upon the concentration of such materials in the products rather than the total amount.

Natural background surveyed

A survey of the natural background over Long Island (New York) was completed in November 1958. The survey which attracted public attention was conducted using a low-flying airplane.

AFL-CIO establishes committee

The AFL-CIO announced in November that it was establishing a committee to study the problems of workers in the nuclear industry. The 14-union Atomic Energy Technical Committee will be chaired by BENJAMIN C. SIGAL, general counsel of the International Union of Electrical Workers.

Health and safety files to open

In the Federal Register of November 27, the AEC proposed to amend its regulations in Part 9, Title 10, C.F.R. to make available for public inspection correspondence with AEC licensees relating to health and safety and to compliance with regulations. Included would be reports to the AEC on radiation accidents.

Radiation engineers certified

Five specialists in "radiation hygiene and hazard control" have been certified by the Board of Trustees of the American Sanitary Engineering Intersociety Board according to an announcement in mid-November 1958. These are among some 979 "sanitary engineers" which it has certified.

Zirconium limits published

The American Industrial Hygiene Association's Hygienic Guides Committee published, in the AIHA Journal for November 1958, a recommended maximum atmospheric concentration for zirconium of 5 mg/m^3 of air. The severity of the hazard was stated as being "low for both acute and chronic exposures" but is based on the possible explosion or fire hazard when in powdered or dust form.

IAEA manual completed

The International Atomic Energy Agency in December 1958 made available its manual on *Safe Handling of Radioisotopes* to be used as the basis of technical rules and regulations among the 69 member states of the Agency. The manual is available from the IAEA in Vienna at $1.00 per copy.

Local concern expressed in Britain

At a meeting on radiation hazards on December 5 in England, local health officers expressed concern over their apparent lack of powers to control radiation health hazards in their locality. They also raised questions as to the hazards to local fire and police departments from emergencies arising in plants using radioactive materials,

Waste panel meets

The *ad hoc* panel of the International Atomic Energy Agency on radioactive waste disposal met in Vienna in December 1958 and decided to start to assemble technical data to form the basis for an international agreement on radioactive waste disposal.

UN Scientific Committee continued

The United Nations General Assembly voted in December 1958 to continue for one additional year its 15-nation Scientific Committee on the Effects of Radiation. The UN Secretary-general also stated that an international scientific conference on the effects of radiation "would be useful" if held under proper auspices.

AEC Approves PRDC permit

The AEC issued an order on December 11 based upon an opinion and initial decision of the Commission to affirm and amend its provisional construction permit for the Power Reactor Development Company power reactor being built at Lagoona Beach, Michigan. The opinions followed a series of hearings and oral arguments since August 1956 when three labor unions requested the AEC to suspend its original provisional permit. The final order has not yet been issued.

Los Alamos worker dies

A 38-year-old laboratory technician at Los Alamos Scientific Laboratory died of radiation damage to the central nervous system on January 1, 1959, some 36 hr after receiving a high exposure in a criticality accident. The estimated dose was reported to have been "in excess of 1000 roentgens" with other estimates ranging up to 8000 roentgens.

Plane test reactor damaged

On December 7 it was reported that a nuclear plane test reactor at Idaho had undergone "a small excursion" on November 18. The AEC stated that the reactor had been removed and examined. It was found that all of the fuel elements and some of the moderator had been damaged by overheating.

Health Physicists in the news

HANSON BLATZ, formerly chief of the radiation branch of AEC's Health and Safety Laboratory, New York Operations Office, is now director of radiation control for the New York City Department of Health. Colonel RALPH M. LECHAUSSE, chief of nuclear medicine, Office of the Air Force Surgeon General, was awarded the air force's Legion of Merit on March 13, 1959 for administration and investigation of possible radiation exposures during 1958 Pacific nuclear tests. RICHARD B. RHODY, a member of the Health Physics Society, died on August 12, 1957. Rhody specialized in radiation detection devices and fast neutron dosimetry associated with localization of brain tumors. Lt. Commander J. H. EBERSOLE, medical officer of the *U.S.S. Seawolf*, and a charter member of the Health Physics Society, received the Gorgas Medal at the Association of Military Surgeons annual meeting for distinguished service in military medicine.

Courses and conferences

In addition to the above training courses, the following courses and conferences may be of interest to health physicists:

Columbia University, Department of Radiology of the College of Physicians and Surgeons: One-year, full-time or two years, half-time, course in "Radiological Physics." The program, leading to the degree of Master of Science, is designed to prepare candidates to carry out all the functions of a physicist in a hospital department of radiology. Prerequisite is a bachelor's degree with a strong background in physics. Fees total about $1200. Contact: Dr. H. H. ROSSI, 630 West 168th Street, New York 32, N.Y.

University of California, "Nuclear Technology Programs." Three courses: nuclear technology study program for engineers—June 22–August 28 at Berkeley; radiological regulation and use for government and public health officials—July 20–July 31 at Berkeley; and nuclear technology survey for executives—July 13–July 17 concurrently at Berkeley and at Los Angeles. Fees are $800, $225, and $150 respectively. Contact: Engineering and Sciences

Extension, Nuclear Technology Programs, Room 100, Building T-11, University of California, Berkeley 4, Calif.

"13th International Congress on Occupational Health," to be held July 25–29, 1960, in New York City at the Waldorf-Astoria. Papers are to cover wide range of occupational health areas and abstracts of papers should be submitted before January 1, 1960 to Dr. IRVING R. TABERSHAW, chairman of the scientific program committee, International Congress on Occupational Health, 375 Park Avenue, New York 22, N.Y. The basic theme of the meeting will be prevention rather than cure.

"Congress on Nuclear Energy," to be held June 16–20, 1959 in Rome, Italy, during the Sixth International Exhibition on Electronics and Atomic Energy, includes an International Symposium on Safety and Site Selection for Nuclear Plants. Contact: ROBERTO LEVI, Comitato Nazionale per le Ricerche Nucleari, Via Belisario 15, Roma.

Health Physics Society items

The Certification Committee of the Society is in the process of forming an "American Board of Health Physics" with details as to health physics certification and requirements to be announced soon. The Society has accepted members in two new classes: associate and affiliate. Plans for the Gatlinburg meeting are well advanced with several innovations planned by the Arrangements and Program Committees. Three American chapters of the Society are being formed in Baltimore-Washington area, in Pennsylvania and in the New York City area. A proposed regional Society chapter in Europe is busily being studied by European members and is extended the best wishes of the Society.

Small accident in Aldermaston

At Britain's Aldermaston Atomic Weapons Research Establishment, a "small release of radioactivity" occurred early in 1959 resulting in no harmful exposure to seven suspected employees.

IAEA meetings

Beginning February 25, 1959 the International Atomic Energy Agency held a three day seminar in Vienna on scanning techniques for determining the distribution of radioisotopes in the human body. IAEA also announced, among other meetings, a conference in Monaco in November on radioactive waste disposal.

National Academy comment

The committee on pathologic effects of atomic radiation of the National Academy of Sciences—National Research Council published a comment in January on the UN Report on the Effects of Atomic Radiation. The NAS committee said, regarding the UN report, that "there are several points on which we are not entirely in agreement" and included among these the impression being created that "leukemia is an *inevitable* result of radiation".

Nuclear aircraft safety group

In January 1959, the AEC and the Air Force announced the establishment of a Life Sciences Working Group for Aircraft Nuclear Propulsion to assist in defining "the human factors problem associated with nuclear propulsion for aircraft and in missiles", and "examine in detail the health and safety characteristics of specific propulsion systems".

ANS sets up Hot Lab Division

In January, the American Nuclear Society announced it had formed a Hot Lab Division from the former independent group.

Work restrictions revealed

In February, the AEC submitted to the Joint Committee on Atomic Energy, a report of restrictions from radiation work of 718 contractor employees from January 1, 1957 through September 30, 1958. Of these, 29 restrictions were in excess of 24 months with 13 employees being "permanently reassigned".

New Piqua site approved

In February, the AEC announced it had approved a new site for the 11,400 kW reactor for Piqua, Ohio, with provisions for additional containment. The new site is across the river from the first proposed site, some 700 ft further away.

Banning of A-ships proposed

A resolution banning nuclear-powered vessels from the Great Lakes was reported as being studied for possible presentation to the International Safety of Life at Sea Convention to be held in London in 1960. The banning would be based on possibility of water contamination in the event of a collision. Many communities depend on the lakes for their water supply.

Sealed sources rules amended

On February 12, the AEC amended its regulation in Part 30 to permit the distribution of certain sealed sources under a general license provided that

the manufacturer of the sources obtains an AEC specific license.

Waste disposal hearings held

In February, the Joint Committee on Atomic Energy held a five-day hearing on radioactive waste disposal. In general the hearings indicated that the waste problem is expensive and still has not been physically or economically solved.

Bone marrow success

Five Yugoslavian scientists who were overexposed to radiation were released from the Curie Hospital in Paris in February and were reported as being apparently cured. N. B. KURVINK of the University of California Veteran's Administration Hospital reported in February that successful results had been obtained by treating irradiated patients with stored bone marrow removed previously from these patients. Kurvink proposed that "the banking of marrow by individuals whose occupations entail hazard of excessive radiation would appear to be a reasonable precaution." In March, the IAEA announced it had sent two staff members to Yugoslavia to study the radiation incident.

Federal control of radiation under review

During February and March the Joint Committee on Atomic Energy published two committee reports, the first on *Selected Materials on Employee Radiation Hazards and Workmen's Compensation* and the second on *Selected Materials on Federal-State Cooperation in the Atomic Energy Field.*

The first document, prepared for hearings held in March, contains several detailed analysis of the occupational radiation protection problem in the U.S. particularly as it may affect the operation of workmen's compensation systems. Organized labor led the fight for a federal workmen's compensation system for all radiation over-exposures. Industrial spokesmen and insurance industry personnel urged continuation and improvement of the present state workmen's compensation laws.

In the second report is included a new AEC proposed amendment to the Atomic Energy Act of 1954 to permit the AEC to withdraw its authority for radiation protection over some sources under AEC supervision in certain states where, by agreement, the state and the AEC felt the state control program is effective to control radiation hazards associated with such sources. The AEC would retain its control over other sources of radiation currently under its supervision.

In the midst of the review of AEC-state control over radiation hazards, the 12 man Radiation Protection Advisory Committee to the Surgeon General of the Public Health Service issued a report on *The Control of Radiation Hazards in the U.S.* recommending that "primary responsibility for the nation's protection from radiation hazards be established in the U.S. Public Health Service". On April 8, Senator LISTER HILL introduced a bill to vest such primary responsibility with the Public Health Service. The Public Health Service approach has since been widely supported by several Congressmen. Hearings on the Hill bill were to be held in May, 1959. Hearings on the AEC proposal were to be held May 19–22, 1959. The possible enactment of either bill could not be determined as of this writing.

NASA site questioned

During the hearings by the JCAE on employee radiation hazards and workman's compensation LEO GOODMAN, atomic energy advisor to the United Automobile Workers requested a full congressional investigation of the granting of an AEC license to the National Aeronautics and Space Administration for its testing reactor at Sandusky, Ohio. GOODMAN questioned the possible "pressure" put on AEC's Advisory Committee on Reactor Safeguards to approve the site in spite of possible questions concerning the safety of the reactor.

Power reactor hearings planned

In March the AEC announced that it will hold public hearings prior to the issuance of construction permits and operating licenses for AEC-owned reactors located at non-AEC installations. The hearings will be part of new procedures to provide a broader public review of safety aspects to include reactors other than those privately-owned.

Radiation hazard studies

The National Academy of Sciences in March announced that it had requested its six committees concerned with the biological effects of radiation to review "the present status of scientific knowledge in this field". During March other groups announcing studies relating to radiation hazards included the Joint Congressional Committee on Atomic Energy (on fallout), a White House initiated study by the Bureau of the Budget with the AEC and Public Health Service (on the proper organization within the Federal government for radiological health) and the opening of the new session of the U.N. Scientific Committee on the Effects of Atomic Radiation.

State control of radiation

States inacting legislation to authorize regulation or registration of hazardous radiation sources during the 1959 legislative sessions include Arkansas, Indiana, Kansas and Tennessee. Such legislation has also been introduced in Arizona, Connecticut, Iowa, Maryland, New Mexico, Pennyslvania and Washington. Some 19 states require registration of radiation sources, as of May 1, 1959.

Health Physics Pergamon Press 1959, Vol. 2. pp. 86. Printed in Northern Ireland

HEALTH PHYSICS SOCIETY NOTICE

The Health Physics Society

Objectives

To promote the work of Health Physics, to improve dissemination of information between individuals in this and related fields, to improve public understanding of radiation hazards and protection and to promote Health Physics as a profession.

Qualifications for membership

The membership of the Society shall consist of members, Associate Members and Affiliates. The Board of Directors may elect to membership in the Society reputable persons and organized groups in accordance with the following principles:

Members shall be persons who have been engaged in some form of health physics activities for at least one year and have been graduated from an accredited school of college grade or have had equivalent training. The Board may recognize one year of experience in health physics as being equivalent to one year of college work, and the prospective member need not be completely versed in all phases of health physics but must be contributing to the general field of health physics at the time he or she applies for membership in the Society. Graduate study in science may be accepted in place of part, or all, of the required period of health physics activity.

Associate members shall be persons who lack the necessary qualifications for election as Members but (a) are engaged in a field of endeavor related to health physics or (b) whose interests in the science would make them desirable Associate Members. Associate Members shall be elected in the same manner as Members but may not hold elective offices or vote except in Local Chapters.

Affiliates shall be organizations approved by the affirmative vote of at least eight (8) voting members of the Board. The privileges of this membership shall be the same as those of individuals except there shall be no voting rights. This class of membership may be withdrawn at any time by a unanimous vote of the Board of Directors.

For Members and Associate Members annual dues are $10.00. Six dollars of these dues are for subscription to the Health Physics journal. For non-subscribing members annual dues are $5.00. The fee for Affiliates is $50.00.

Application blanks for membership and information concerning membership may be obtained from the membership committee. Completed applications should be returned to the Chairman, Admissions Committee, Mr. W. E. Nolan, University of California, Radiation Laboratory, P. O. Box 808, Livermore, California.

Health Physics Pergamon Press 1959. Vol. 2, pp. 87–99. Printed in Northern Ireland

FOURTH ANNUAL MEETING OF THE HEALTH PHYSICS SOCIETY

18–20 June 1959 at Gatlinburg, Tennessee

Abstracts and titles of papers presented at the meeting

Session A: Monitoring and Control of Environmental Radiation Hazards.

Atomic weapons accidents—alpha emitters dispersal and decontamination.* Floyd W. Wilcox and John S. Coogan, Radiological Safety Division, Reynolds Electrical and Engineering Company, Inc., Mercury, Nevada.

One of the most probable hazards associated with accidents involving nuclear weapons is the plutonium. It may appear as a dispersal of small particles resulting from impact, explosion or fumes if a fire occurs. This contamination could not expect to occur in ideal conditions since an accident does not respect time or place. It is conceivable that a fire or explosion in a plutonium processing plant would result in plutonium contamination similar to that from a nuclear weapon accident.

At the conclusion of a testing program at the Nevada Test Site, large land areas were left contaminated with alpha emitting material in a manner similar to that expected from an atomic weapon accident. This report presents dispersal patterns and concentration levels for the alpha emitting material and information concerning airborne activity prior to, during and following decontamination. Decontamination methods and efficiencies are discussed, with techniques utilized to prevent personnel exposure and spread of contaminant.

Adsorption of xenon in an activated charcoal column. H. P. Cantelow, Health Chemistry, Radiation Laboratory, University of California, Berkeley California.

Performance characteristics of two activated charcoal columns at room temperature in separating fission product xenon from an air stream were investigated by installing each column in the exhaust from an enclosure in which irradiated slugs were dissolved. Break-through curves are presented and the variation in xenon concentration within the columns is examined. Theoretical treatments of adsorption columns in the literature are found to agree well with the experimental data. Performance of the columns is evaluated in terms of "concentration factor" and number of effective theoretical plates.

High efficiency filter pitfalls. M. D. Thaxter, Health Chemistry, Lawrence Radiation Laboratory, University of California, Berkeley, California.

Users of "high efficiency" filters for cleaning off-gases are advised against adopting an attitude of unquestioning faith in their efficiency. Field data supporting this advice are presented. Pleas for not only further development but also more critical field evaluation are offered.

Health physics program for the U.S. Army Ionizing Radiation Center, Lathrop, California. John L. Donovan, Nuclear Sciences and Engineering Department, Curtiss-Wright Corporation, Quehanna, Pennsylvania.

The utilization of ionizing radiation to preserve foods holds great promise as a very important application of atomic energy for peaceful uses. The success of this new method of food preservation depends upon the effective development of safe and efficient production processes at an economical cost. It is for the attainment of this objective that the U.S. Army Ionizing Radiation Center is being planned. No such irradiation facility has been designed or constructed before.

The stringent health and safety specifications for a food processing facility dictate highly specialized health physics programs. Two sources of ionizing radiations will be used. The High Intensity Food Irradiator (HIFI) will employ over two million curies of cobalt-60. The Linear Accelerator (Linac) will produce electrons with energies up to 32 MeV. Each facility is capable of processing 3000 lbs of food/hr with doses of 2 million rad, or a capacity of 3 Mrad tons/hr.

Each of these facilities is the largest unit of its kind ever built. Each has its own health physics problems. Successful design and operation of these units requires a broad application of health physics procedures.

The special problems and the final solution of the preoperational radiation background survey program will be described. This is probably the first background survey program to use gamma scintillation counting techniques exclusively. Information will be provided for

* Work was performed under the auspices of the U.S. Atomic Energy Commission.

both specific isotope analysis and general gamma radiation background. Thus the added sensitivity and selectivity of scintillation counting is used for providing information to guard against contamination from the cobalt used in HIFI, or activity from positron emitters originating from gamma-neutron reactions at the Linac facility.

The personnel exposure control program will be outlined. With the high dose rates present in the irradiation facilities, human residence time is less than 1 min. The control program to prevent entry into the facilities during production operation will be outlined.

The exacting contamination control procedures required for purity of the food products imposes other severe control problems. Highly sensitive monitors are used to check on the storage pool water, the air exhausted from the facilities and the food packages leaving the exposure areas to re-enter the food plants. Display meters in the control room and audible alarms will warn the operators of the presence of any contamination.

Fission product uptake by trees in the vicinity of ORNL waste pits. S. I. AUERBACH, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

Monitoring of core wells within and around the margin of the ORNL waste pit area shows that Ru^{106}, Co^{60}, and Sb^{125} migrate laterally. Bark, twig, leaf, and ground litter samples were taken from seventeen species of trees growing in this marginal area, during the periods April–July in 1957 and 1958. Objectives were to determine the specific activity of Ru^{106} and the accumulation of it and other fission products in tree components and ground litter. The distribution of contaminated trees coincided with the pattern of dispersion of radionuclides adduced from core well analyses. Concentrations of Ru^{106}, the most abundant fission product in tree material ranged from 10^{-5} to 10^{-1} μc/g dry weight. Year-to-year comparisons gave no evidence of accumulation of Ru^{106} but a seasonal increase in concentration was found in the leaves which appeared to be positively correlated with seasonal maturation. Other fission products found were Co^{60}, Cs^{137}, and Zr^{95}-Nb^{95} with concentrations from 10^{-6} to 10^{-4} μc/g dry weight. Autoradiographs made of leaves taken at varying distances from the pits showed evidence of radioactive particulates on leaf surfaces. The number of particulates per unit area appeared to be inversely related to distance from the pits which tends to support the hypothesis that Cs^{137} and Zr^{95}–Nb^{95} contamination is airborne rather than ground-dispersed.

Maintenance and field performance of gas-flow alpha survey instruments under extreme environmental conditions.† HAROLD C. WOODS and FREDERICK M. CRANDALL, Radiological Safety Division, Reynolds Electrical and Engineering Company, Inc., Mercury, Nevada.

This paper describes the performance of a popular alpha survey instrument in year-around outdoor use under the extreme climatic conditions found at the Nevada Test Site. Maintenance problems produced or aggravated by these environmental variations are discussed.

Alpha monitoring is performed at the Nevada Test Site, under field conditions, at altitudes varying from approximately 3000–7500 ft above sea level. These altitude changes cause shifts in the alpha and beta sensitivity of monitoring equipment. Such shifts in sensitivity can also result from the rapid variations in relative humidity and temperature which occur when an instrument is taken from open-air areas into underground locations. Additional problems are presented by static electricity accumulation due to low relative humidities, by air-borne sand and dust, and by seasonal variations in temperature from 0° to 115°F.

While the gas-flow principle of alpha detection simplifies these problems considerably, the final solution to many of them was found to lie in modification of accepted maintenance and calibration procedures.

Fallout and natural background in the San Francisco Bay area. LLOYD D. STEPHENS, H. WADE PATTERSON and ALAN R. SMITH, Lawrence Radiation Laboratory, Berkeley.

On or about 20 March 1958 a significant increase in the background radiation levels was noticed at the University of California Lawrence Radiation Laboratory. It was soon determined that this increase was associated with radioactive material deposited during a series of rainstorms. The Health Physics department began a series of measurements in the San Francisco Bay area in order to contribute to the assessment of the extent and amount of the deposited radioactivity.

The instrument used for the field surveys was a NaI(Tl) crystal 3 in. long × 3 in. dia. and viewed by a DuMont K1197 phototube, the output of which drives a count-rate meter.

This same crystal was used for gamma-ray pulse-height analysis in conjunction with a 50-channel pulse-height analyzer. Sample counting is done inside of a 4 in. thick lead shield.

In making this survey, the points were chosen to be as evenly spaced and as representative of altitude, rainfall and rural and urban areas as possible.

One area, an area of dense fern growth, showed the highest radiation levels and it was decided to use this fern for pulse-height analysis. Samples were ashed and then analyzed. The results of this analysis did bear out the belief that this was fallout produced radiation. The prominent peaks at 0.15, 0.49 and 0.75 MeV are characteristic of fallout spectra of this age. The 1.62 MeV La^{140} peak was also present.

The results of the several surveys enabled us to establish a value for the natural background radiation for these areas as none was available prior to the deposition of the fallout.

By the use of gamma-ray spectroscopy in conjunction with a gamma-ray dose field survey, it is possible to measure changes in the radiation level due to fallout that in magnitude do not exceed 10 per cent of the natural

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

† Work was performed under the auspices of the U.S. Atomic Energy Commission.

background. A continuing program is being carried out both in the field surveys and the spectroscopy.

Halogen tube remote area monitoring system. H. A. BROWN and J. V. ROGERS, Reactor Monitoring Center, Tracerlab, Inc., Richmond, California.

After analyzing radiation monitoring requirements and studying various systems, Tracerlab has evolved a new area monitor which offers the advantages of no zero drift, five-decade range and only two monthly calibration adjustments. A small halogen quenched GM detector is used in a unique circuit that will cover the range background to 50 r/hr. At radiation levels below $\frac{1}{10}$ r/hr the detector operates as a normal pulse GM detector while at high intensities it acts as an ion chamber. The circuitry utilizes transistors in the probe, and produces a logarithmic response. The alarm features permit reading radiation level from any channel even after exceeding the alarm level. Remote meters for calibration purposes or remote indication can be plugged directly into the sealed probes.

Organization of a Regional Radiological Assistance Team. PERCY GRIFFITHS, Health and Safety Division, U.S. Atomic Energy Commission, Idaho Falls, Idaho.

In January 1958 a Radiological Assistance Plan was established by the General Manager of the U.S. Atomic Energy Commission. As a result, the Nation, including its possessions, was divided into Regions for this purpose, each Region with the capability to assist and advise local authorities with problems encountered in incidents involving radioactive materials. This paper describes the organization and function of a Regional Team.

Problems associated with the shipment of radioactive materials. PERCY GRIFFITHS, Health and Safety Division, U. S. Atomic Energy Commission, Idaho Falls, Idaho.

Many problems incidental to the protection of the Public accompany the shipment of radioactive materials, The precipitous increase in shipments of radioactive materials over the past decade has intensified these problems. This presentation briefly covers the various regulations controlling the shipment of these materials, the various exemptions, and field experience. A number of accidents and incidents are reviewed and illustrated in detail.

Microwave radiation hazards. W. W. MUMFORD and M. M. WEISS, Bell Telephone Laboratories, Whippany, New Jersey.

A review of the published literature on the exposure of animals to microwave radiation indicates that the chief hazard appears to be due to the heating effect resulting from the absorption of energy. The interpretation of extensive experimental data on small fur bearing animals has led to the establishment of recommended exposure limits. These limits are discussed from the standpoint of the probable safety factor involved for total immersion of the human being in the micro-wave field and for localized exposure confined to particular portions of the anatomy.

Precautionary measures are discussed and a method for estimating the distance from radiating antenna is presented.

Session B: Radiation Physics.

Investigation of windows and shields for neutron point sources. HOWARD J. BROWNE, J. A. KAUFMANN, N. B. GARDEN, Health Chemistry, Lawrence Radiation Laboratory, University of California, Berkeley, California.

An empirical approach for the evaluation of shielding materials for macro-chemical manipulations of spontaneously fissioning heavy elements (curium and californium) has revealed interesting comparisons. High-density metal-halide solutions were compared with lead glass and with rare-earth glass for use as shielding windows. Laminated shields of lead-paraffin and uranium-paraffin were compared with water and with paraffin for shielding walls. Measurement of samples of the pure shielding materials compared favorably with published data, where available. Some interesting results were achieved when the materials tested were used in combination. On the basis of the figures obtained dose attenuation factors were computed for various thicknesses of the materials tested.

"Sticky" collisions as a mechanism for three body stabilization of O_2^{-*}. G. S. HURST, Health Physics Division, Oak Ridge National Laboratory,† Oak Ridge, Tennessee.

The cross-section for capture of low energy electrons to O_2 in mixtures of O_2 and N_2 depends on the pressure in the following way:

$$\alpha = Af_1P + Bf_2P + Cf_1Pf_2P$$

where α is the coefficient of attachment of electrons to O_2, f_1P is the oxygen pressure, f_2P is the N_2 pressure, and A, B, and C are empirical constants. The large value of the constant C is a matter of considerable interest since it indicates the importance of three body collisions. This large rate of three body stabilization of O_2^{-*} now appears to be interpretable in terms of "sticky" collisions. In this model O_2^{-*} (unstable against electron emission) collides with N_2 to form the loosely bound complex $(O_2^{-*}\text{–}N_2)$. The lifetime of the $(O_2^{-*}\text{–}N_2)$ complex against molecular dissociation may be shown to be quite long (5×10^{-9} sec) compared to molecule–molecule collision times (10^{-13} sec). This lifetime is also much longer than the lifetime for electron emission which, by using the experimental constant C, may be estimated to be about 5×10^{-11} sec, compared to the Bloch-Bradbury estimate of 10^{-10} sec. Collisions of O_2 with the $(O_2^{-*}\text{–}N_2)$ complex within the time 5×10^{-11} sec can lead to stable O_2^- by virtue of a highly efficient process in which energy is transferred to O_2.

An improved method for measuring electron attachment. T. E. BORTNER, Health Physics Division, Oak Ridge National Laboratory,† Oak Ridge, Tennessee.

The earlier method[1] used to study the process of

† Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

[1] T. E. BORTNER and G. S. HURST, *Health Physics* **1**, 39 (1958).

electron capture to O_2 employed an alpha source to produce the free electrons. This led to two limitations: (1) the number of electrons per pulse was limited to about 2×10^5 which in turn limited the amount of oxygen that could be used, and (2) a non-attaching gas at a pressure greater than that required to stop the alpha particles in the chamber had to be used. Neither of these difficulties appears in a new apparatus which employs a photoelectric surface and a pulsed ultraviolet light source for the generation of the pulses of electrons. In this method two identical ionization chambers are used, each receiving light from the same ultraviolet flash tube. One chamber contains a non-attaching gas, the other contains a mixture of the same non-attaching gas and O_2. The ratio of the pulse heights furnishes data from which the attachment coefficient is calculated. The double chamber arrangement eliminates the uncertainty caused by fluctuation of the light output. The pulse heights are determined with an oscillator-ramp circuit in which the number of pulses recorded on a scaler is proportional to pulse height.

Attachment of electrons in O_2–CO_2 and O_2–CH_4 mixtures. L. B. O'KELLY, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

The attachment of low energy electrons in small amounts of oxygen mixed with methane and carbon dioxide has been studied over the pressure range of 250 mm Hg to 800 mm Hg and for E/P values from 0.1 to 3.0. The apparatus, which uses a pulsed ion chamber, has been described previously by BORTNER and HURST.[1] A considerable difference in the sensitivity of the two gases to oxygen contamination was found. Concentrations of oxygen as high as 30 mm Hg in 370 mm Hg of methane reduced the pulse height approximately one-half while 0.35 mm Hg in 400 mm Hg of carbon dioxide reduced the pulse height to about one-third of its value in pure carbon dioxide. A definite dependence of the attachment on both the oxygen pressure and the pressure of the non-attaching gas was found. The attachment cross section decreased as the electron energy increased in accord with other investigations in this energy range.

Measurement of electron flux in tissue-like media bombarded by X-rays. H. H. HUBBELL, JR., R. A. FINSTON, and R. D. BIRKHOFF, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

The study of various factors involved in RBE values reported previously[2] was continued with further measurements of the yield of electrons from parallel plate chambers bombarded by X-rays in vacuum. For graphite plates the yield in esu cm^{-2} rad^{-1} absorbed in air, when multiplied by the ratio of the absorption coefficient for X-rays in air to that in carbon was a constant, 3.6×10^{-4} esu cm^{-2} rad^{-1} absorbed in graphite independent of X-ray energy for heavily filtered X-rays of effective energies ranging from 45 keV to 200 keV. The energy spectra of the electrons were measured with considerably increased accuracy, sensitivity and electron energy range by an a.c. method.[1] Signals of frequencies 300 and 324 counts/sec and of equal amplitude modulate the d.c. plate voltage. The beat frequency amplitude is proportional to the second derivative of the current voltage curve. The first derivative was obtained by integration and both were used to determine the flux. The spectral distribution of the number of electrons in graphite does not follow the previously found inverse square dependence on energy for electron energies above about 6 V. Results for tissue equivalent plastic plates were essentially the same as those for graphite.

Measurement of stopping power by calorimetric methods—I. Calorimeter design and scattering effects. R. D. BIRKHOFF, F. KALIL,* W. G. STONE,† and H. H. HUBBELL, JR., Health Physics Division, Oak Ridge National Laboratory,‡ Oak Ridge, Tennessee.

Electrons of energy E pass through a thin conducting foil, losing energy ΔE, and are collected in a Faraday cup. Two identical sets of thermocouples are in thermal contact with the ring which supports the foil, one measuring temperature of the foil-ring system, the other providing heating or cooling to the system by the Peltier effect. The system is described by

$$n/2i^2R - n\pi Ti + I\Delta E + 2nL(T_0 - T) = mc\dot{T}$$

where n is the number of Peltier thermocouples of resistance R carrying current i with Peltier coefficient π and absolute temperature T. I is the beam current, L the heat conducted per thermocouple per degree, T_0 the reference junction temperature, and mc the heat capacity of the system. In the rate-of-drift method, $\dot{T}$ is measured with $i = 0$. In the null method, $T = T_0$ and $\dot{T} = 0$. Calibration in either method is obtained by reducing the beam energy until electrons are completely absorbed in the foil ($\Delta E = E$). Singly-scattered electrons striking the ring result in an energy E being delivered to the system (instead of ΔE), whereas multiple scattering results in path lengths exceeding the foil thickness.

Measurement of stopping power by calorimetric methods—II. Foil preparation and results for Al, Cu, and Au. R. M. JOHNSON, P. L. ZIEMER, F. KALIL,* and R. D. BIRKHOFF, Health Physics Division, Oak Ridge National Laboratory,‡ Oak Ridge, Tennessee.

Foils for stopping power measurements must be thin to minimize scattering, uniform, electrically and thermally conducting, and free of pin holes. Self-supporting foils satisfying these criteria were made by evaporating from one filament a soluble compound (Victawet, $FeCl_3$, NaCl,

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

[1] T. E. BORTNER and G. S. HURST, *Health Physics* **1**, 39 (1958).

[2] R. D. BIRKHOFF, D. R. NELSON, H. H. HUBBELL, JR. and R. H. RITCHIE, *Health Physics* **1**, 219 (1958); ORNL-2521.

* Now at Glenn L. Martin Co., Baltimore, Maryland.

† Now at Redstone Arsenal, Huntsville, Alabama.

‡ Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

[1] R. A. FINSTON, W. G. STONE, H. H. HUBBELL, JR. and R. D. BIRKHOFF, Bull. SE Section American Physical Society (April 1959).

were floated off the slide in a suitable solvent (usually water). Foils were weighed on a Cahn Electrobalance, modified by using storage batteries, precision Helipots, a K-2 Potentiometer and precision resistor to read the meter current, a microscope to observe the pointer, and a small a.c. current through the meter to overcome friction. The sensitivity was improved from ± 5 μg to ± 0.1 μg. The rate of drift method, with Al foils of thickness 50.3, 107.8, and 172.3 μg/cm^2 and a Cu foil of 191.8 μg/cm^2 yielded stopping power data in good agreement with the Bethe theory for electron energies from 10 to 125 keV. However, data from a 201.6 μg/cm^2 Au foil gave a stopping power 7.4 times higher than theory because singly scattered electrons struck the ring supporting the foil.

On the stopping power of ions in tissue. J. NEUFELD and W. S. SNYDER, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

In order to calculate the RBE dose delivered to tissue it is often essential to know the linear energy transfer of protons, alpha particles, and of carbon, nitrogen and oxygen ions. At the present time there is no exact theory for calculating the stopping power in those energy ranges that are of particular interest in dosimetry and related fields. In a recent publication,[1] SNYDER and NEUFELD have computed the stopping power of various ions in tissue by a method suggested by KNIPP and TELLER.[2] The above calculations[3] are based on experimental data that have since been superseded by more exact values. The present calculation involves three simplifying assumptions: (1) the mass stopping power is the same for a solid medium as for a gas of the same chemical composition; (2) the stopping power is the sum of the stopping powers for each element of the medium; and (3) the stopping power is the product of the mean square charge of the particle and a "specific stopping power". The mean square charge is estimated by extrapolation and interpolation of experimental data and the specific stopping power is calculated from experimental values for protons.

Fast neutron spectra and dose rate calculations. J. B. EGGEN, R. L. FRENCH and A. REETZ, JR., Convair, A Division of General Dynamics Corporation, Fort Worth, Texas.

This paper describes a method and its application for calculating fast neutron spectra and dose rates about the Lid Tank Facility source plate, the Bulk Shielding Reactor, and the Aircraft Shield Test Reactor. The method may be applied to any reactor whose shield is composed largely of hydrogenous materials. Dose rates and spectra calculated by the method are useful in shield design work and in the analysis of experiments performed in reactor radiation fields.

The method is based upon fast neutron differential energy spectra and dose rates calculated by the Nuclear Development Corporation of America for a point isotropic fission source in infinite media. These data represent a moments method solution of the Boltzmann transport equation for water and hydrocarbons. By representing a reactor core as an array of point isotropic sources and assuming that any material along the line-of-sight between a point source and a point detector may be represented by an equivalent thickness of water or hydrocarbon, the fast neutron attenuation through any material may be determined by use of the results for these reference materials. The equivalent thicknesses of a non-hydrogenous material are determined from the ratio of the effective removal cross sections.

Results from dose rate calculations by this method agree with measured BSR and LTSF data within 10 per cent for centerline distances between 40 and 120 cm. Calculated and measured spectra for the BSR and the ASTR compare favorably, although the ASTR calculation involves penetration of considerable amounts of non-hydrogenous materials.

Low level alpha counting. F. J. BRADLEY, Office of Radiation Safety, The Ohio State University, Columbus, Ohio.

The vibrating reed electrometer in the rate of charge mode of operation is a sensitive detector of alpha-rays. Using an argon gas flushed chamber, an alpha background of 0.10 alpha disintegrations per minute has been obtained with stainless steel construction. This low background enables one to detect alpha activity in the range 1 $\mu\mu$c.

Comparison is made with the proportional counting, liquid scintillation and track counting techniques. The lower limit for age determinations is estimated to be 10^{14} years for alpha radiation.

A fast neutron pulse reactor for health physics and biomedical research. H. P. YOCKEY, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

A fast neutron pulse reactor is proposed to supply a pulsed or steady neutron source for research in neutron dosimetry, radiobiology, medical therapy, and other fields. The design concept is based on the "Godiva" reactor at the Pajarito site in Los Alamos Scientific Laboratory. The neutron yield of this reactor is stress limited. This limitation may be substantially reduced by using two ideas due to Stratton, namely, constructing the reactor of many separate pieces and using an alloy of uranium. Two designs for the reactor core are a nest of concentric cylinders and a bundle of hexagonal rods. The fraction of the total burst energy which appears as mechanical energy available to do damage is drastically reduced. A further advantage is that enhanced yield is limited by thermal and metallurgical considerations. A further improvement possibly can be made by employing alloys of U with 20–30 at. % Mo and 1–4% Zr, Ti, or

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

[1] W. S. SNYDER and J. NEUFELD, *Rad. Res.* **6**, 67 (1957).
[2] J. K. KNIPP and E. TELLER, *Phys. Rev.* **59**, 661 (1941).
[3] W. S. SNYDER and J. NEUFELD, ORNL-1083 (November, 1951).

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

Nb which have much better mechanical and metallurgical properties than pure U. It is expected that the proposed reactor may be operated at a yield of 3–5 $\times$ 10^{16} fissions and possibly as high as 10^{17} fissions per pulse.

Session C: Waste Disposal.

Application of the maximum permissible concentration of mixed fission products to waste processing. ALAN A. JARRETT, Atomics International, Canoga Park, California.

Apart from the utilization of natural decay to reduce the activity of waste fission products, the most general method of treating radioactive waste has been to concentrate and store the bulk of active material and, if possible, to dilute and disperse the remainder to the environment. The concept usually employed to measure the efficiency of a treatment process has been the decrease in radioactive concentration or decontamination factor. The reduction of biological toxicity, however, is more suitable than the decontamination factor since both the reduction in activity and changes in toxicity (MPC) are taken into account. For this reason, both the composition (MPC) and activity level were used to calculate the changing biological toxicity of decaying mixed fission products. A typical waste treatment process will be used to illustrate the application of this concept from both economic and radiological hazard aspects.

The influence of clay mineral structures on cesium sorption. T. TAMURA, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

Clays were investigated for their ability to retain cesium from high-level waste. Illite was found to be most efficient in preferentially sorbing cesium from sodium-neutralized Purex waste (6 M). For example, when 2.5 ml of waste solution containing 0.25 mg of cesium was mixed with 1 g of illite, 0.15 mg (60%) was sorbed. The mineral vermiculite sorbed 3 times as much cesium when the spacing between the 001 planes was decreased as compared with the natural state. When altered in this way, the vermiculite held approximately 1½ times as much cesium as did the same weight of illite.

The capacity of the local Conasauga shale for cesium was measured. The natural shale sorbed 0.10 mg of cesium/g; following the treatment which induced collapse of the 001 planes of vermiculite, the shale sorbed 0.25 mg/g. The sorption of cesium can be explained on the basis of structural parameters of clay minerals.

Sorption of ORNL intermediate-level wastes by Conasauga shale. D. G. JACOBS and W. J. LACY, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

Disposal of intermediate-level wastes at ORNL involves the discharge of the waste into a series of three 1-million gal pits. These pits are excavated in the Conasauga shale formation. The beta activity of the waste is about 1.5 $\times$ 10^5 c/min per ml, which is too great for direct release to the uncontrolled environment. The solution is essentially 0.29 N $NaNO_3$ and is alkaline (pH 10.6). Conasauga shale has been found to be effective for removal of cesium from a waste stream. The saturation capacity of the shale was determined by passing increments of simulated and actual waste solutions through known weights of the shale. The Cs/Na distribution factor was 200. This corresponds to a saturation capacity of 350 μc of Cs/g shale for this waste solution. The shape of the sorption front is of importance in determining the potential longevity of the pits. This front is dependent on the reaction rate, specificity of bonding, and efficiency of contact. Sorbed cesium is very difficult to remove by percolating ground water through the shale. Because cesium is held at exchange sites, removal is increased by increasing the concentration of cations in the leaching solution. Conasauga shale is 85 per cent effective in the removal of gross activity from a mixed fission product waste. However, based on MPC_w values, the shale is 98 per cent effective in removing high yield, hazardous radionuclides.

Laboratory work on certain aspects of the deep well disposal problem. W. J. LACY,* Health Physics Division, Oak Ridge National Laboratory,† Oak Ridge, Tennessee.

If there is sorption near the point of injection of the radioactive waste into a well, the concentration of activity per cubic foot may present a heat production problem due to radioactive decay. Cesium is a long-life fission product known to be easily sorbed by clays. The amount of cesium sorbed in laboratory experiments was found to be a function of several variables: (1) concentration of cesium in the waste, (2) concentration of other cations in the waste, (3) amount and nature of the clay present, (4) composition and amounts of other minerals in the formation, and (5) hydrogen ion concentration of the waste.

Illite sorbed 3.5 mg of cesium/10 g of clay from a simulated 6 M neutralized Purex-type waste solution, while kaolinite sorbed 1.1 mg/10 g and halloysite less than 0.5 mg of cesium. Increasing the sodium ion concentration from 0.5 M to 6.0 M decreased the amount of cesium sorbed from 1.02 to 0.39 mg/g of clay. Decreasing the cesium concentration from 100 mg/l. to 10 mg/l. resulted in a decrease in the amount of cesium sorbed from 0.35 mg/g to 0.11 mg/g of clay. A mixture of small amounts of limestone with illite reduced the uptake of cesium from neutralized Purex waste. About three times as much cesium was removed from a sodium neutralized solution as from an acid solution.

The use of competing ions, like sodium and calcium, or the adjustment of the Na/Cs ratio can control the

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

* Now at Office of Civil and Defense Mobilization, Battle Creek, Michigan.

† Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

sorption of cesium within the range where localized concentration of heat will not represent a hazard to waste disposal through deep well injection.

Preliminary studies of the disposal of radioactive wastes in natural salt formations. L. HEMPHILL, W. J. BOEGLY, L. BRADSHAW, F. M. EMPSON, and F. L. PARKER, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

The National Research Council (Earth Sciences Division) recommended that natural salt formations be considered for the long-term storage or disposal of radioactive wastes. This recommendation was based on the (1) impermeability, (2) plasticity, and (3) widespread distribution of rock salt. In salt disposal, the principal considerations are: (1) the thermal problem, and (2) chemical compatibility. The calculated maximum temperature rise in a sphere 20 ft in diameter containing reactor fuel reprocess wastes aged for one, six, and eleven years indicate that cavities of this size can store these wastes without exceeding 200°C, the temperature at which plastic flow becomes significant. Laboratory studies of the chemical interaction between synthetic Purex waste and rock salt show that the solubility of salt, as well as the gas evolved, can be controlled by proper choice of acidity, temperature, and ionic concentration.

Treatment of large volume, low-level waste at ORNL. I. Design features of plant and evaluation of performance. K. E. COWSER and R. J. MORTON, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

At Oak Ridge National Laboratory, the collection, treatment, and disposal of large volume (0.5–0.7 million gal/day), low-level radioactive liquid waste is accomplished through a process waste water system. Based on laboratory and pilot plant studies, horizontal flow units employing the lime-soda softening process were selected for treatment purposes. Construction of the plant was completed August 1957 at a cost of $250,000. The plant employs gravimetric chemical feeding, flash mixing, coagulation, and sedimentation, and has a capacity of 500,000 gal/day. If needed for increased efficiency, the recirculation of plant effluent, the return of preformed crystals (sludge), and repeated precipitation are possible. An 800,000 gal equalization basin is provided to minimize fluctuations in chemical composition of the influent wastes. Operating experiences has shown that the plant is capable of removing 85–90 per cent of the radioactive strontium and rare earths by the softening process. About 85 per cent of the radioactive cesium is removed by the addition of 150 p.p.m. of clay. This waste treatment plant is unique in that it is the first to be designed and operated specifically for the removal of radiostrontium from large volumes of low-level waste.

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

Treatment of large volume, low-level waste at ORNL. II. An application of response surface methodology for optimizing treatment efficiency. D. A. GARDINER and K. E. COWSER, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

The treatment plant was designed to allow the addition of various amounts and combinations of lime, soda ash, clay, returned sludge, and calcium chloride. To determine the optimum combination of the five variables, a plan of laboratory experimentation was devised and partially completed to measure the removal of cesium and strontium as a function of these variables. The design of the experiment chosen was one in which the response surfaces of cesium and strontium removal could be estimated by a quadratic equation in the five treatment variables. A test of the efficiency of the quadratic model showed that the deviations of the observed cesium responses from the fitted model were not significant at the 30 per cent level. However, the deviations of the observed strontium responses from the model were significant at the 0.1 per cent level. The effect of each variable, separately and in combination, and the characteristics of the cesium and strontium response surfaces were estimated and their significances determined.

Dispersal of radiocontaminants in streams. F. L. PARKER, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

The physical, chemical, and biological properties of a stream frequently can ameliorate the effects of a sudden or continuous release of radioactive materials to such a stream. The sudden release can be studied from a physical viewpoint only, and has been tested by "slug" tracer methods. The chronic release must be examined more especially from the biological viewpoint and has been studied by the buildup in the muds, biota, and water. A simplified theoretical equation which fits the experimental results is obtained for the sudden release:

$$C = \frac{M}{\sqrt{4\pi A \alpha^2 t}} \exp\left[-\frac{(x - vt)^2}{4\alpha^2 t} - (N + \lambda)t\right],$$

where C and M are the concentration and mass of the contaminant, A the cross sectional area of the stream, α^2 the eddy diffusion coefficient, t the time after release of activity, v the mean velocity of the stream, N the river uptake coefficient, and λ the nuclear decay constant. Attempts to set up such a model for the chronic case have not been successful.

Surface water monitoring following sea disposal operations. R. E. ALEXANDER, Health Physics Unit, Atomics International, Canoga Park, Calif.

Atomics International obtains disposal for radioactive waste by sea burial to a chemical disposal area approximately 30 miles off the Southern California coast. Although no bulk disposal operations are conducted,

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

inquiries by various California agencies to the AEC have created a need for data to indicate whether these disposal operations significantly raise the radioactive material concentrations in the surface waters of the disposal area. A procedure for supplying the data has been effected, and is described in this report.

Surface water samples are obtained in the disposal area before and after each operation. A background sample is taken approximately 10 miles from the area on the same day. These samples are analyzed for mixed fission products and uranium, the most likely contaminants. Mixed fission products are co-precipitated with $CaCO_3$, using $NaCO_3$ as the precipitating agent. Quantitative measurements are obtained by beta counting. Uranium is complexed as diethyldithiocarbamate and extracted into chloroform. Quantitative measurements are obtained by polarography. Results are analyzed statistically to determine with 75% confidence whether an increase has occurred.

The problems of high salts concentration, variable potassium and nuclear test debris concentrations, and the variable presence of plankton are discussed. The error in the mixed fission product co-precipitation technique as caused by mixed fission product age is shown by gamma spectra analysis to be much smaller than might be expected, even though $Cs^{137} + Ba^{137}$ is not recovered, and Nb^{95} and $Ru^{106} + Rh^{106}$ are only partially recovered.

Session D: Radiation Dosimetry.

Automatic Nuclear Emulsion Scanner. STANLEY BECKER, Airborne Instruments Laboratory, Mineola, N. Y.

An Automatic Nuclear Emulsion Scanner has been designed and constructed to scan the film in personnel radiation monitoring badges. The Scanner detects a portion of proton-recoil tracks in the film emulsion caused by incident neutron flux. The scanning speed, at present, is comparable with that of the visual reader providing about the same statistics.

The Scanner uses a special closed-circuit TV camera system to view the magnified image of the dark-field illuminated nuclear emulsion. The information out of the camera is analyzed in a computer which utilizes two ultrasonic delay lines for signal storage. The computer discriminates against single grains, clumped grains, and unusual artifacts in the emulsion. Tracks are detected on the basis of their contrast against their background, and their projected length in the emulsion.

Field evaluation tests at Brookhaven National Laboratory indicate a linear track count as a function of neutron flux. The spectral response of the Scanner shows an enhancement of track count at the higher energies. In addition, the Scanner track count is fail-safe for gamma exposures of up to about 1.5 r.

Photographic dosimetry at total exposure levels below about 20 mr. MARGARETE EHRLICH and WILLIAM L. MCLAUGHLIN, Radiation Physics Laboratory, National Bureau of Standards.

There is an increasing awareness of the need for dosimetry at total exposure levels down to those of natural background. Photographic dosimetry at these levels is difficult, because of limitations in the sensitivity of commercially available emulsions and because of instabilities and calibration inaccuracies encountered in the sensitometric procedures.

Among the methods that suggest themselves as suited to aid in surmounting these difficulties are: Increase in total exposure times, resulting in an increased density at a given exposure rate; the use of several film samples of the same type in stacks, resulting in an increase in the ratio of total useful film density to background density; the use of film in contact with phosphors, whose ultraviolet emission adds to the photographic effect due to the high-energy photons; improvement in sensitometric procedures. The advantages and limitations of these methods will be discussed; special attention will be given the problems of latent-image fading and reciprocity failure.

Silver-phosphate glass dosimetry: fluorescence method. J. A. AUXIER and W. T. THORNTON, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

Small glass rods (1×6 mm) and a fluorescence measuring device have been used in a study of the dosimetric characteristics of silver bearing metaphosphate glass.[1–5] The commercially available system† was analyzed for consistency and reproducibility, photon energy dependence from about 20 to 1250 keV, fast neutron response for several energies, and thermal neutron response. Several techniques for encapsulation of the glass to correct for photon energy dependence were evaluated. In general, gamma correction for increased response in the photoelectric energy region utilized the techniques commonly employed for film. With simultaneous calibration, necessitated by lack of an adequate fluorescence standard, measurements of gamma-ray doses above 10 r are reproducible to $\pm 5\%$. Measurements at the ORNL 2.5-MeV Van de Graaff yielded the following upper limits for fast neutron response relative to gamma radiation response: 1.5 MeV, 0.45%; 1 MeV, 0.6%; 0.5 MeV, 0.45%. A response equivalent to that of 1 r is produced in the glass by 5×10^9 thermal neutrons/cm^2.

Radiation monitoring utilizing average current from halogen quenched GM tubes. JAMES M. GARNER, JR. and R. LOUIS BRADSHAW, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

Simple instruments utilizing the average GM tube current and having either linear or logarithmic response to radiation intensity can be made. The type of response and sensitivity is governed by choice of GM tube type and the circuit parameters of capacitance, resistance, and

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

† Bausch and Lomb Optical Co., Rochester, N.Y.

1 N. J. KREIDL and G. E. BLAIR, *Nucleonics* **14,** No. 3, 82 (1956).
2 J. H. SCHULMAN *et al.*, *J. Appl. Phys.* **2,** 1479 (1951).
3 J. H. SCHULMAN and H. W. ETZEL, *Science* **118,** 184 (1953).
4 H. W. ETZEL, R. D. KIRK and J. H. SCHULMAN, *Ra-Det* **8,** No. 2, 49 (1955).
5 A. L. RIEGERT, H. E. JOHNS and J. W. T. SPINKS, *Nucleonics* **14,** No. 11, 134 (1956).

voltage. The effects of these parameters on eleven types of halogen-quenched GM tubes have been investigated.

Full-scale ranges as low as 1 mr/hr and as high as 20 r/hr are practical in instruments which do not require vacuum tubes or transistors. Greater sensitivity may be obtained by using two or more GM tubes in parallel, the new type microammeters, or one stage of current amplification.

Trichloroethylene-saturated water and tetrachloroethylene chemical dosimeter systems. Neutron response. D. G. Ott, W. H. Schweitzer, J. A. Sayeg and P. S. Harris, Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico.

Chlorinated hydrocarbon chemical dosimeter systems are capable of measuring gamma dose from about 5 to 2,000,000 r using photometric evaluation methods. The tetrachloroethylene, two-phase system has only slight response to fast neutrons which makes it valuable for measuring gamma dose in mixed radiation fields. The trichloroethylene-saturated water, one-phase system (which performs very satisfactorily as a gamma dosimeter) has a considerable response to fast neutron dosimetry. Attempts have been made to use this type to measure neutron dose. Such dosimeters would be valuable for biological experiments (particularly for depth–dose measurements) if energy dependence is not a factor.

Responses of both types of dosimeters to neutrons of various energies have been determined by exposure to known doses of monoenergetic neutrons at 1, 2, 4, 6, 8 and 14 MeV and from fission neutrons. Compared to response to Co^{60} gamma radiation, neutron response of the two-phase C_2Cl_4 system has been found to vary from about 2 per cent at 1 MeV to 16 per cent at 14 MeV. Approximately 4 per cent response was obtained from fission neutrons with the aqueous indicator phase present, and about 1 per cent when anhydrous C_2Cl_4 was used. Response of the one-phase, C_2HCl_3-saturated water system varied from about 15 per cent at 1 MeV to 56 per cent at 14 MeV, and was about 35 per cent for fission neutrons.

One rad of thermal neutrons produced a response equivalent to approximately 3.5 rads of gamma radiation in the C_2HCl_3–H_2O dosimeters and to 4.5 rads with the C_2Cl_4 system. This effect of thermal neutrons is usually eliminated by shielding the dosimeters with $\frac{1}{2}$ in. of lithium.

From the results it can be concluded that the neutron response of the C_2Cl_4 system is sufficiently low and constant to allow good measurements of gamma dose in mixed radiation fields. Neutron energy dependence of the aqueous one-phase system. however, limits its use for measurements of fast neutron dose.

An automatic film badge processing machine. Leo F. Kocher, Hanford Laboratories, General Electric Company, Richland, Washington.

A machine has been designed and constructed which will automatically process the Hanford personnel film badge. The badge machine will X-ray code the film packet in the badge for identification, magnetically release the badge lock, disassemble the badge, remove the exposed film packet, place a new film packet in the badge, and relock the badge. The operations for processing of the badges (electrical and pneumatic) are controlled by a variable speed pneumatic timer. Badges and new film packets are fed into the machine by magazines which are independently indexed and controlled from the timer.

Instrumentation consists of double and single acting air cylinders, pneumatic indexing table, control valves, vacuum pump, X-ray machine, electrical drive motors, micro-switches, solenoids, and an electromagnet.

Improved inspection throughout and considerable cost savings are realized by the use of the automatic badge processing machine. The machine was completed for routine operation in February, 1959.

Some limitations of film meters as personnel radiation dosimeters. Harold W. Henry and Hugh F. Henry, Oak Ridge Gaseous Diffusion Plant, Union Carbide Corporation, Oak Ridge, Tennessee.

The effect of some of the factors which, as practical considerations in personnel monitoring, influence the accuracy of radiation dose determinations by film meters were checked with radium and cobalt sources and "phantoms" of a urea-sucrose solution. From these measurements, it was concluded that uncertainties in any dose determination were easily possible by the magnitudes indicated as follows:

(1) The attenuation due to an individual's body—300–500%;
(2) Backscatter from an individual's body—10–20%;
(3) Simple reversal of the film packet in the badge—30%;
(4) Difference in backscatter due to energy (open window only)—10%.

In additional experiments wherein the badges were placed inside the phantoms which were then rotated in the unidirectional radiation field, it was noted that the radiation doses recorded by badges at any position inside the rotating phantom were approximately equal, but these doses were only about 75 per cent of that indicated by a badge on the phantom surface. In addition, this surface badge gave a reading approximately 60 per cent that of a badge on the front surface of a similar stationary phantom. The actual experimental results and their implications, as they apply to routine and emergency monitoring, are briefly discussed.

Mixed radiation dosimetry of a plutonium-beryllium neutron source. D. C. Lawrence, Atomics International, Canoga Park, Calif.

The dose rate delivered to an assumed "standard tissue" by the fast neutron and gamma radiation from a standard one curie plutonium-beryllium neutron source was determined by measurements with ethylene filled polyethylene and CO_2 filled teflon ionization chambers. The resulting ionization currents as measured with a calibrated vibrating reed electrometer were used to obtain a measure of both the gamma and fast neutron component

of the emitted radiation. The tissue dose rate due to the fast neutrons at 1 cm from this source was found to be 1.83 rep/hr. The gamma component was found to contribute 0.19 rep/hr. From the neutron dose rate the emission rate of the source was calculated to be 1.63×10^6 neutrons/sec-curie. A complete discussion of the neutron and gamma spectra of the emitted radiation from this source and some aspects of ionization chamber design are also included in this report.

Improvements in personnel metering procedures at the National Reactor Testing Station. F. V. CIPPERLEY and W. P. GAMMILL, U. S. Atomic Energy Commission, Health and Safety Division, Idaho Falls, Idaho.

The rapid growth of the NRTS and the increased interest in occupational radiation exposure experience has pointed up the need for improved techniques in external exposure determination and more efficient methods of exposure record tabulation.

The improved procedures at the NRTS, utilizing a modified Hanford type multiple filter film badge and IBM tabulating equipment are described as well as a method of plotting film densities behind the various filters for energy determinations and differentiation of beta and soft gamma.

Of particular interest is the mechanization of film reading, record production and compilation of statistical reports.

Session E: Internal Dose.

The activity of chain decaying nuclides collected at a constant rate. R. C. BARRALL, Mare Island Naval Shipyard, Vallejo, California.

Chain decaying nuclides present the health physicist with special problems of sampling and dosimetry. Radon-222 and thoron (radon-220) are probably the most frequently encountered since they are found in normal environmental air where their presence may delay or prevent accurate measurement of aerosol activity due to other nuclides.

In addition these nuclides are of direct interest as:

(1) Possible sources of hazard in certain industrial situations.
(2) Contributors to man's normal environmental exposure.
(3) Dosimetry problems in experimental work and in calculating MPC values for these nuclides.

When chain-decaying nuclides are collected from an equilibrium mixture at a constant rate, a steady state (but not equilibrium) is eventually reached. The relative activity of the various members of the decay chain is a complex function of the decay constants and time. A precise knowledge of the activity of each of the successive daughters in a chain when collected at a constant rate may be of significance in engineering applications and is directly applicable to internal dosimetry and sampling processes.

By integration and successive substitution in the well known Bateman equations, one may develop rigid equations expressing the activity of successive daughter nuclides continuously for all values of time during and after collection.

A method of calibrating detectors for chain-decaying nuclides utilizing simultaneous solution of the Bateman equations is indicated.

Public health hazard of bone-seeking fission products. N. IRVING SAX, Radiologic Health Laboratory, New York State Department of Health, Albany, N. Y.

This paper presents a discussion of the public health significance of the radiologic contamination of population exposure vectors such as food, air, and water by mixed fission products. An attempt is made to assign public health hazard priorities to the bone-seeking components of these mixed fission products as opposed to the so-called gross beta activity. Analytical technics and instrumentation for the determination of several sources of population exposure to radiation are discussed, as are possible lines for future development of this work.

The epidemiological follow-up of the New Jersey radium dial painters. LESTER A. BARRER, New Jersey State Department of Health, West Orange, N. J.

The New Jersey State Department of Health has entered into contract with the U.S. Atomic Energy Commission through the Division of Biology and Medicine for a study concerned with the relationship between the concentration of radium and mesothorium retained in the body for long periods of time and the effects of this body burden upon the individual. The study has been in operation for approximately one year.

The persons to be studied were those who used radium and mesothorium in their work approximately 35–45 years ago. There are approximately 1000 individuals in the metropolitan New Jersey and New York area. This group constitutes the largest group in the United States, possibly in the world.

The study is being conducted in a series of steps:

(1) Identification of and search for the present location of all the former employees of the radium industry.
(2) A personal interview and follow-up to obtain a family and occupational history.
(3) Medical, dental and laboratory and radiographic studies to obtain information concerning the past and current history of the individual.
(4) Radiological studies to obtain information about the identification of and the concentration of radioactive materials retained in the individual.
(5) Analysis of the data collected to relate body burden of radium and mesothorium with the medical history of the individual.
(6) Statistical studies to relate levels of body burden of radium and mesothorium with the incidence of pathological findings for the entire group.

At present, the identification of the names of approximately 500 radium workers has been made. Over 100 living individuals, each with an estimated body burden greater than 0.01 μg of radium, have been found and are

assisting the New Jersey State Department of Health in this study. Medical and radiological studies are being conducted on these individuals. It is planned that each of these cases will undergo a whole body count in the new whole body counter being constructed by the Atomic Energy Commission and the Institute of Industrial Medicine at Bellevue Medical Center.

We are still looking for and finding additional living radium cases.

Metabolism of Zn^{65} in mice, rats, dogs, monkeys and man.* C. R. RICHMOND and W. H. LANGHAM, Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico.

The ubiquitous occurrence of zinc in tissues and its function as a prosthetic group in enzymes concerned with proteolysis and both cellular and gaseous respiration emphasize its prominent role in divergent metabolic processes. Zinc may also participate in the functioning of myelogenous bone marrow and erythropoiesis.

Although Zn^{65} is not a product of nuclear fission, it has been identified as a contaminant in people and food from the Hanford area, in a group of cyclotron workers at this Laboratory, in human beings and marine organisms from the Marshall Islands, and in fallout.

Large-volume liquid scintillation counters and a 9 × 6 in. sodium iodide crystal gamma-ray spectrometer were employed to detect *in vivo* time changes in the whole body retention following acute administration of oral doses (≤ 1 μc) of Zn^{65} to mice, rats, dogs, monkeys, and man. Distribution of Zn^{65} in the tissues of rats and the effect of chronic administration on retention in dogs were also investigated.

Results show that at least three exponential rates contribute to the loss of radiozinc from the bodies, or individual tissues, of the animals studied. Bone and pelt accumulated larger fractions of the administered dose and exhibited longer turnover times than did other tissues. A possible relation between species and uptake from the gut was observed, the larger animals being more efficient in absorbing zinc.

The data clearly indicate an effective half-time of more than 100 days for humans and suggest an increase for the most recently recommended values for f_1, f_2, and f_w.

Study of internal contamination in the Marshallese people. STANTON H. COHN and JAMES S. ROBERTSON, Brookhaven National Laboratory, Upton, L. I., N. Y.

Studies of the body burdens of those Marshallese who accidentally received internal contamination in March, 1954, were first made by radiochemical analyses of urine samples. In 1957 the body burdens of several of those Rongelapese were measured by Miller and Marinelli at the whole body counting facility which they developed at Argonne National Laboratory. A similar facility was subsequently developed for an on-the-spot check of the Marshallese by Brookhaven National Laboratory.

In 1958 a steel room 5 ft × 5 ft × 4¼ ft with 4 in. walls was shipped from Brookhaven Laboratory to the Marshall Islands. Within this shield, a sodium-iodide crystal was used to detect internally deposited isotopes. Signal pulses were analyzed with the aid of a 100-channel pulse analyzer.

The findings from the 1958 group survey indicate that the principal internally deposited isotopes are Cs^{137} and Zn^{65}, as was also determined by Miller and Marinelli in their study on the selected group. The levels of the isotopes measured in 1958, however, are several times the levels present in 1957. The data suggest that the increasing body burden is the result of ingestion of contaminated food. During the interval between the 1957 and 1958 studies, the Rongelapese, who had been moved to Majuro following the accident, were returned to Rongelap.

Experience with the technique of whole body counting shows that it is a rapid and sensitive means for determining the level of contamination in a population.

The concentration and distribution of elements in the human body. ISABEL H. TIPTON, Department of Physics, The University of Tennessee and Health Physics Division, Oak Ridge National Laboratory.*

During the past eight years autopsy specimens of some 30 tissues from over 250 individuals from the United States and 150 from other parts of the world have been analyzed for trace elements using methods of emission spectrography, neutron activation, and flame photometry. This paper will report the results of analyses of "normal" subjects—that is, victims of instantaneous accidental death—and compare these values with those for tissues from subjects who died with various diseases. (The concentrations of certain elements in specific tissues increase with age, and geographical variations of certain element configurations exist).

***In vivo* gamma counting as a measurement of uranium in the human lung.** R. E. COFIELD, Y-12 Plant,* Oak Ridge, Tennessee.

A direct, *in vivo* method is reported for the measurement of uranium within the human lung, utilizing the detection by scintillation spectroscopy of the gamma radiation accompanying alpha decay of the uranium. The *in vivo* gamma counting facility includes a low background counting chamber with 8 in. thick iron walls, a 9 in. dia. × 4 in. thick NaI scintillation detector, and a 256-channel pulse height analyzer. To achieve a quantitative estimate of uranium concentration, the normal, non-uranium gamma activity at the characteristic uranium gamma energies must be subtracted from the gross spectrum of the subject. In addition to the method of subtraction of the spectrum of a second person, not exposed to uranium, a second technique using spectral prediction equations derived from the gamma spectra of persons not exposed to uranium has been developed. Calibrations equating the excess count rate to uranium lung burden have been derived from measurements of samples in phantoms

* This work was done under the auspices of the U.S. Atomic Energy Commission.

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

simulating the human chest. The 186 keV gamma line from U^{235} is the most suitable for the measurement, but if secular equilibrium is assumed, the 90 keV lines of the uranium daughters may be used in exposure cases when the relative abundance of U^{235} is low. The ratio of the 90 keV line to the 186 keV line may be used as an indication of U-235 abundance. The limits of the measurement (x, 0.05) in a counting period of 30 min are about 130 μg of U^{235} using the 186 keV line and about 10 mg of natural uranium using the 90 keV lines.

The method has been used to measure uranium lung burden in several cases, on which data and some comparisons with excretion analysis results are presented.

Exposure of embalmers and physicians by radioactive cadavers. J. S. LAUGHLIN, S. J. VACIRCA and J. F. DUPLISSEY, Department of Physics, Memorial Center, New York, N. Y.

For radioactive cadavers whose surface exposure dose rate was more than 30 mr/hr the policy was initiated at Memorial Center a few years ago of requiring embalmers to perform their work at the Memorial morgue under our supervision. During each supervised embalming, the undertaker and his assistants wore two calibrated pocket dosimeters each. In the last year 16 cadavers with radioactive iridium were embalmed under our supervision and exposures ranging up to 40 mr were measured. On the basis of these measurements and general considerations with respect to the handling of cadavers, it appears advisable that radiation safety precautions be recommended when the surface exposure rate exceeds a specified amount (such as 100 mr/hr), rather than on the basis of the radioactivity content of the body.

In the case of other cadavers which contained radioactive solutions (such as radioactive phosphate), the embalming apparatus necessarily became contaminated. Although the absolute amounts of radioactivity may not be large, because of the affinity of phosphorus compounds for the skin, it is important that the apparatus be carefully decontaminated. It appears advisable in the case of radioactive solutions, even though the radioactivity content of the body may be small, that radiation safety recommendations with regard to the handling and cleaning of apparatus be required.

This report will discuss these measurements and recommendations, as well as determinations of the exposure of physicians during autopsy procedures.

Session F: Criticality Hazards and Reactor Safety.

Criticality accident applications of the ORNL badge dosimeter. E. D. GUPTON, D. M. DAVIS and J. C. HART, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee.

Recent nuclear accidents have emphasized the need for and the requirements of a personal meter that would be effective in accurately determining, in a relatively short time, the neutron and gamma radiation dose received by personnel in the vicinity of such accidents. The Oak Ridge National Laboratory badge meter has been modified by the addition of materials which permit measurement of moderate to high level neutron exposures and which permit immediate knowledge of those persons who may have received medically significant doses of gamma or neutron radiation. These modifications supplement the previously incorporated desirable features of the badge, such as provisions for wide range beta-gamma dosimetry and low level neutron dosimetry. A bare gold foil, a cadmium covered gold foil and a pellet of sulfur are the badge components to determine moderate to high level neutron doses. An indium foil and a liquid chemical dosimeter are the components which permit immediate segregation of those who may have received medically significant neutron and/or gamma exposures. Provision is made for inclusion of silver phosphate glass rods to supplement the gamma dose measurements. This meter, in conjunction with Neutron Threshold Detector Units of the type reported by HURST *et al.*,[1] would provide information which is of the utmost importance following accidental high level radiation exposures to personnel.

Problems of personnel monitoring at a criticality incident. HUGH F. HENRY, Oak Ridge Gaseous Diffusion Plant, Union Carbide Corporation, Oak Ridge, Tennessee.

Radiation exposure determinations at a criticality incident are complicated by the fact that the radiation from such an incident is essentially unidirectional and the effect of the human body in attenuating both gamma and neutron radiation and in moderating and scattering neutrons complicates the use of any simple monitoring device. Analysis of some experimental data with film meters for gamma monitoring and indium foils as neutron monitors will be presented, and this indicates that these factors can easily produce uncertainties in dose determination by factors of at least three or more. It is concluded that it would be very difficult to devise a simple personnel monitoring device which would be both acceptable for general use and would also provide sufficiently accurate data for rapid and dependable analysis; thus, accurate monitoring may comprise a comparatively complicated system. A proposed system which includes a simple personnel device, body fluid analysis, and the use of area-type neutron and gamma dosimeters will be described in principal.

Administrative control of criticality hazards at Pratt & Whitney Aircraft—Canel. WILLIAM F. PATTON, Connecticut Operations, Canel, Middletown, Connecticut.

Some radiological safety studies of conventional reactor housing structures. W. P. KUNKEL, Atomics International, Canoga Park, California.

The objective of this paper is to examine the atmospheric containment characteristics of a conventional reactor housing structure.

* Operated by the Union Carbide Corporation for the U.S. Atomic Energy Commission.

[1] G. S. HURST *et al.*, *Rev. Sci. Instrum.* **27,** (1956).

Containment requirements are based on the radiation exposures to the public in surrounding areas as a consequence of a reactor accident. (The accidents considered for this paper are those which release fission products to the reactor building structure without damaging the integrity of the structure by other means).

An estimate is made of the environmental hazard (from leaking reactor structures) in terms of the building leak tightness and the reactor characteristics. The modifying effects of volume source, elevated discharge and multiple barrier containment are discussed.

A research reactor, critical facility, and power reactor are compared as to containment criteria for comparable site-boundary exposure.

The physical mechanisms of contaminated atmosphere release from building structures are evaluated in terms of geometry effects; and pressure differentials and various leakage rates resulting from the induced pressure differentials of the environmental factors of temperature, density, wind, and barometric pressure are compared.

Results indicate the adequacy of conventional reactor housing structures for safe atmospheric containment of many reactor-site configurations.

Pre-operational health physics indoctrination for reactor personnel. LIONEL LEWIS, Combustion Engineering, Inc., Windsor, Connecticut.

The author considers it particularly desirable to instruct reactor personnel in health physics theory and to inform them of radiation safety procedures and requirements prior to reactor completion and startup. In accord with this view a training program was established for reactor operating, maintenance, installation and test groups and other interested persons. This paper describes and outlines the content of the Health Physics Indoctrination Lectures and Supervisor's Health Physics Discussions that were given prior to completion and operation of a Submarine Prototype Power Reactor Facility.

The preparation of the lecture material, the arrangements for the lectures, the method of presentation and the results derived are also discussed.

Health physics coverage for startup of the Argonne Low Power Reactor (ALPR). E. D. GRAHAM and P. G. STODDART, Argonne National Laboratory, Idaho Division, Idaho Falls, Idaho.

The Argonne Low Power Reactor (ALPR) is located at the National Reactor Testing Station and was brought into operation in 1958. ALPR is a boiling water reactor with an enriched uranium core and is cooled and moderated with light water. Thermal heat production is 3.0 MW, from which 300 kW of electric power and 200 kW of usable thermal space heat are obtained. ALPR was designed for use in remote areas and all components may be airlifted by standard operational military aircraft.

The health physics aspects of assembly, startup, and initial power operation, including a 500 hr continuous run at full rated power, are summarized. Conditions common to closed-cycle boiling water reactors are discussed, together with conditions peculiar to ALPR. Personnel exposure data are tabulated and diagrams show typical radiation background levels.

Health Physics aspects of Borax IV operation with defective fuel. E. D. GRAHAM and P. G. STODDART, Argonne National Laboratory, Idaho Division, Idaho Falls, Idaho.

Argonne National Laboratory operators of the BORAX IV at the National Reactor Testing Station in Idaho had, for some time, planned an operational experiment to be conducted with a direct cycle boiling reactor power plant with a known defect in fuel cladding.

On March 11 and 12, 1958 this opportunity was presented and the experiment conducted with the reactor operating at low power (2.4 MW). Although radiological data collection was not considered as the prime reason for this experiment, it was felt that certain data of this type must be collected and reviewed as the experiment progressed. It was also felt that these data should be compiled and made available to interested personnel outside the organization.

Radiological information gained from this experiment consisted of:

(1) The extent of area contamination downwind of the plant.
(2) In-plant contamination levels to which operating personnel were exposed.
(3) Isotopic identification of contamination released.
(4) In-plant radiation levels to which operating personnel were exposed.
(5) Identification of fission gases released to the atmosphere through the air ejector stack and to the water.

Safety aspects of nuclear powered aircraft. CARL C. GAMERTSFELDER, General Electric Company, ANPD, Cincinnati, Ohio.

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SYMPOSIUM ON TECHNICAL METHODS IN HEALTH PHYSICS

25–28 May 1959 at the Danish Atomic Energy Center, Risø

A SYMPOSIUM on Technical Methods in Health Physics was held at the Danish Atomic Energy Center, Risø, Denmark on May 25–28, 1959. This symposium was sponsored by the European Nuclear Energy Agency of the Organization for European Economic Cooperation. It was attended by official delegates and speakers from 15 countries and representatives from a number of the international organizations. Dr. NIELS BOHR, Chairman of the Danish Atomic Energy Commission, acted as honorary chairman. This symposium was a great success and marks the beginning of such Health Physics symposia in Europe. The complete papers of this conference are being published by the European Nuclear Energy Agency of OEEC, Paris and should be available by the fall of 1959. In the meantime the *Health Physics Journal* is pleased to offer its readers the opportunity to read the following abstracts of the papers given at this symposium.

Welcome Address by Prof. NIELS BOHR

It is a great pleasure and honour, on behalf of the Danish Atomic Energy Commission, to extend a most hearty welcome to all the participants in this Symposium, arranged under the auspices of O.E.E.C. by the European Nuclear Energy Agency and with the collaboration of the U.S. Atomic Energy Commission.

We in Denmark are grateful and proud that so many eminent experts should come to this meeting—experts with special knowledge and experience in the now so important problems of protection against radiation—protection both for the public and for those engaged in research and development projects.

Nuclear development in this country is as yet only in a preparatory stage. But we are grateful, through our membership of O.E.E.C., to have the opportunity to collaborate in the important joint undertakings like Eurochemic in Belgium, the Halden project in Norway, and the recently established "Dragon" project at Winfrith Heath in Great Britain.

I shall not go into further details. I just want these few words of welcome to express the warmest wishes for the success of the Symposium. I am confident that it will materially contribute to knowledge of these most important and vital problems, and am especially sure that it will be of great benefit to the work of our own group here at Risø. In conclusion I sincerely hope that all our guests from abroad will have some pleasant days in Denmark.

Introductory Remarks by Prof. Dr. R. M. SIEVERT

Your Excellency Professor NIELS BOHR, Ladies and Gentlemen.

I would like on behalf of the Symposium to thank Professor BOHR for his kind words of welcome.

It was with great pleasure that I received the invitation to come to the Risø Symposium and I would like to express my sincere thanks to the European Nuclear Energy Agency as well as the Danish Atomic Energy Commission for the opportunity which has been given to me to take part in this meeting.

I congratulate the Organizers of the Symposium on the most valuable initiative which they have taken in arranging the first European Symposium of this special nature with its very interesting and attractive program.

May I also say how much we all appreciate that this first meeting is being held in Denmark, the country of NIELS BOHR to whom we owe so much.

I am anxious from the very beginning to state that my introductory remarks at this session reflect my personal opinions and that these may not necessarily coincide with the opinions of my colleagues in the ICRP or in the United Nations Radiation Committee.

We live in a period when radiation hazards have become a subject of general interest and, I would

like to add, have become a problem which for many persons, even some scientists, is approached with an *a priori* feeling as being something more threatening to mankind than most other existing perils in the world. The present situation can, however, as far as the radiation protection specialists are concerned, also be characterized as a period of reflection and transition, indicating perhaps that many of our present ideas on radiation effects, and particularly on their significance to mankind, may need revision.

Let us have a glance into the historical background. Already in the first years after the discovery of X-rays and radioactive substances, a relatively large number of those who worked closely with ionizing radiations developed severe injuries—and within the next few decades radiation had caused the premature death of some 100 radiologists. As a reaction against this, radiation protection gradually improved over the following period of about 30 years and the recommended maximum permissible doses of radiation became set at relatively low levels. The growing knowledge of the effects of radiation on the haematopoietic system was taken as a further reason for extreme cautiousness. It is also important to state that there were in general no major practical difficulties in the way of increasing protection in medical radiology to the levels of the recommendations given by the predecessor of the ICRP—the International X-Ray and Radium Protection Committee—and the costs of such technical protection arrangements were not frightening.

When the production and use of radioisotopes began and progress in the nuclear energy field greatly increased the sources of ionizing radiations, there was a general demand to keep the possibilities of receiving harmful radiation doses at an extremely low level. The previous experience from the early use of X-rays, and partly also the new kind of exposures being met with in work with large quantities of radionuclides, made this attitude well justified. Another reason was that the occurrence of even a very few radiation injuries would have badly influenced public opinion as regards the new field of human enterprise introduced by atomic energy work. The maximum permissible levels of radiation were thus further decreased.

At present in atomic energy plants there is a similar position to that generally occurring in the medical use of ionizing radiation, that is to say there are no major drawbacks which accompany the addition of the radiation protection necessary to ensure that the doses received by the personnel are well below the maximum permissible levels recommended by the ICRP. I feel that the occupational exposure of personnel to ionizing radiation is today so small that the radiation risks involved are far below most of the risks which occur in other types of work.

It is impossible to predict whether this fortunate situation will last, but it may be that we shall soon come across an increasing number of occasions when individuals and groups of persons will be exposed to many different sources, occupationally and otherwise.

If the same maximum permissible doses are used for *all* types of exposure, these doses will have to be set at the minimum value with respect to those particular procedures where radiation exposure can be easily avoided. This would lead to unnecessary restrictions of other procedures which might be of such considerable benefit that the establishment of a higher maximum permissible level for them would be justified. Thus we will probably soon have to face the question of either increasing the maximum permissible levels and accepting an increased risk of harmful effects, or of limiting the production of ionizing radiation or—which is most probable—both. In order to keep a sound balance between benefit and harm we might therefore need different maximum permissible levels, dependent upon the aim of the radiological work in which persons are exposed to radiation.

The risks of harmful effects of radiation are often dealt with as a problem different in principle from other risks involved in ordinary living.

There are some attributes of ionizing radiation which actually make its biological effects different from those of other agents. One is the fact that radiation effects are often long delayed—even so as to affect later generations—another is that any part of the body can be affected or damaged due to an exposure received in a fraction of a second as well as due to a dose accumulated over many years. This makes it necessary, for instance, to proceed slowly, even if we find indications which would appear to allow higher maximum permissible radiation levels than the present ones. I would, however, like to stress that in the long run this does not place radiation effects in a unique position as compared with other agents, and certainly does not justify the disregard of the general principle of a reasonable balance between the possibilities of benefit and harm in relation to radiation safety. I believe that it is necessary today to point out that the risk of radiation injuries must be considered as something which is, in principle, not very much different from other risks occurring in a modern community.

There is, however, a complication which is often not taken into consideration.

To the delayed and cumulative nature of the effects of ionizing radiation we have to add, as I have already said, the consideration of the infinite number

of different ways in which radiation can reach the human body. From radiotherapy we know that for arge quantities of radiation there is, besides the dose-effect relationship, a marked dose-rate-effect relationship. To elucidate the importance of this complication even under small doses I will only draw attention to the recent investigations by the RUSSELLS and by CARTER on genetic effects, which seems to show that mutations induced by radiation are not dose-rate-independent as was earlier thought. This indication that the use of "extrapolation", for the determining of the effects of small doses from the actual investigations at high dose levels, is invalid, has made the determination of the hazards of small doses even more difficult to assess than before. As you all know such extrapolations have been the main basis for the estimations of the genetical radiation hazards at low radiation levels. A similar kind of extrapolation is sometimes made to arrive at the number of radiation induced cases of leukemia or the reduction of the life span in a population subject to low level irradiation.

All these extrapolations have been made in order to arrive at figures indicating the maximum possible frequency of injurious effects.

Let me take an example to show how dubious this kind of approach to the radiation safety problem might be. Assume that a person is exposed continuously to an irradiation, in addition to the irradiation from natural sources, amounting to 30 per cent of the average natural level. This means an extra dose rate of about 10 mμr/sec. Assume that another person is exposed to the same weekly dose but over a period of only 7 sec in the week—then the latter person receives radiation at the dose rate of 0.1 mr/sec.

The additional exposure of the first person could also be obtained by changing his living conditions within normal limits. If he is a sailor and changes over to work on land living in a wooden house he will increase his exposure by an amount of the magnitude of 30 per cent over his average natural level while at sea, as regards both dose and dose rate. If he lives in a wooden house and moves to an ordinary red brick house he will have about the same increase in exposure.

The person, however, who receives this same additional weekly dose concentrated into 7 sec is exposed to the same 30 per cent increased dose, but at a dose rate which is several thousand times higher than the normal dose rate to which the human body is usually exposed.

We have thus to consider if there is the same likelihood of harmful effects due to the radiation received in two such extreme cases, that is;

(a) the organism receives a dose in addition to the natural background by a continuous irradiation within the limits of the variations of the natural dose rate and

(b) the organism receives the same dose during a short interval of time, but with a dose rate which is several powers of ten higher than can be found in nature except perhaps in some very rare cases.

We have at present some, but not sufficient, indications to be able to answer this question; but we must nevertheless in our practical protection work try to make use of a reasonable hypothesis. This is what we can call trying to be "foreseeing", and in a field where we lack knowledge to such an extent as we do here, on the effect of small radiation quantities, we must try to be foreseeing. A sound principle seems thus to assume that there might be an increased probability of a biological effect at low levels of radiation—especially in those cases where the additional irradiation differs substantially in any respect from what can be regarded as being within the normal variations of the natural irradiation conditions of the living organism.

There is one aspect especially, which we have to take into account as a consequence of this principle, namely in the collecting of data on irradiated personnel: we must determine not only the total dose but also the dose rates, even if they can only be roughly estimated. I would therefore at this symposium like to urge that the dose rate be specifically considered as an important factor and recommend also that the dose rate be noted in the health surveys of atomic energy plants.

Finally I would like to emphasize the necessity of a close cooperation between countries where individuals and populations are exposed to ionizing radiation substantially exceeding the average natural level. Radiation accidents are few and diseases and abnormalities caused by radiation are rare. We therefore need the experience from many countries to be able, as soon as possible, to arrive at figures showing the real risks caused by radiation. To be able to carry out worldwide investigations of radiation effects on man we must of necessity also be able to know the doses and dose rates in an adequate and standardized manner, so as to make comparisons possible.

This Symposium constitutes one of the most valuable ways in which information and discussion of methods and instruments for such purposes can take place. I am firmly convinced that this Symposium will be of the greatest importance for the future work on the protection of man against ionizing radiations.

Abstracts of papers presented at the symposium

Session I: *Fundamental Scientific Principles.* Chairman: Prof. R. M. SIVERT (Sweden).

Recent advances in the knowledge of radiation injury and the basic standards of radiation protection. A. FORSSBERG, Karolinska Institutet, Stockholm, Sweden.

It is proposed to restrict this survey mainly to experiences from mammalian radiobiology. Certain findings in the fields of radiation chemistry and cellular radiobiology are, however, useful for an understanding of the basic mechanism of radiation effects, particularly the appearance of gene mutations, and will be briefly considered.

It has been maintained that the output of gene mutations is proportional to dose down to the smallest amounts of radiation. There is now good evidence that irradiation, protracted over fairly long periods of time, causes considerably less mutations in mice than from linearity expectation. We know also that mutations are not definitely manifest in the moment they are formed, but that in a period of sometimes up to 1–2 hr postirradiation there are experimental possibilities decreasing the mutation yield in micro-organisms.

A variety of physiological effects arise after heavy doses of whole body irradiation of animals. Detailed analyses show that organs and tissues differ greatly in radio-sensitivity; the ultimate death of heavily irradiated animals is therefore the integrated result of a complicated and only partially understood chain of reactions. Yet the doses needed to kill 50 per cent of a population of various mammals show remarkably small variation, indicating a mainly uniform radiation response.

Recent reports on reinvestigations of human populations exposed to heavy radiation will also be briefly considered (Hiroshima, Nagasaki, Marshall-islanders).

Effects at low dose levels, either from repeated single exposures or from protracted continuous radiation, require particular consideration. Apart from genetic effects we have mainly to reckon with production of neoplastic diseases (tumours), particularly leukemia, as well as shortening of life span, "aging". Various opinions have been expressed on the significance of these lesions and difficulties arise when animal experiments are evaluated and conclusions drawn for man. This is particularly true when we approach doses close to background radiation. The phenomenon of radiation-induced aging is moreover poorly understood.

Spectacular progress has been made in the fields of radiation protection and therapy by biological-chemical procedures. Quite a few substances of diverse chemical constitution, particularly SH-compounds, afford substantial protection against radiation death when given shortly before the exposure; in the last years it has been found that a number of pharmacologically very active chemicals also give a good protection. These latter findings seem to open new aspects on the mechanism of radiation effects and their protection. From the procedures which have been found to be of therapeutical value after the reception of a high dose, bone marrow cell transplantation stands as the most important principle.

The scientific basis for maximum permissible limits of internal contamination. W. S. SNYDER, Oak Ridge National Laboratory, Oak Ridge, Tennessee, U.S.A.

Maximum limits of internal contamination depend on the conditions of exposure and also on the permissible level of risk. They are based preferably on direct observation of the accumulation of the radionuclide in the body and of its effects on the body under similar conditions of exposure. For most radionuclides and for long-term chronic exposure, such data are not available, and the limit is set in terms of a maximum permissible dose rate to the various organs of the body. Such a limit is based on effects observed when the organ is irradiated at a similar level, either by another radionuclide or by external radiation, and on general experience with radiation effects at low levels of exposure. Some long-term data on the effect of radium in bone is known, and this is used to set the permissible level for radium and, by comparison of dose levels, for other radionuclides

that deposit in bone. Clinical experience with radiotherapy has provided considerable direct evidence of the effects of internal and external radiation on man. Laboratory experiments with animals have been the source of much relevant data but generally demand interpretation and revision for application to chronic exposure of man. The development of wholebody counters has already provided some relevant data, particularly on retention of various isotopes and such counters will doubtlessly be tools of increasing importance in this work. Much of the data from the above sources is at dose levels considerably above those where the effects are marginal or difficult to detect and thus it must be reinterpreted. The background level of exposure may be considered to give a lower bound for a permissible level of exposure even in the case of exposure of the whole population.

Session II: *Measurement and Control of Radiation and Contamination in Working Areas.* Chairman: Dr. P. RECHT (Euratom).

Basic criteria in the control of air and surface contamination. D. E. BARNES, Health Physics, Aldermaston, Berkshire, U.K.

1. *Permissible dose to tissue*

Objective in control of air and surface contamination is to ensure that exposure received by any organ or tissue shall be within the permissible limits. There are international agreed values for persons occupationally exposed and for the general public—reasons why these differ—values for farm animals. Summary of maximum permissible doses from which all other criteria are derived.

2. *Concentration in air*

Maximum permissible concentrations in air have been derived by the ICRP. Brief survey of methods used—effect of particle size and solubility—behaviour of particles in respiratory tract—effects of energy of radiation, uptake factors, size of critical organ, effective half-life—some typical values.

3. *Surface contamination*

Routes by which surface contamination may become a hazard—direct irradiation—inhalation—ingestion—absorption through wounds or intact skin—from grass to animals. In many of these cases the factors required are not well established—discussion of experimental data and of assumption which may be made in the absence of data. Derivation of levels on the basis of these consumptions—determination of the case which sets the limit—suggested values for permissible levels.

Some main figures indicated during the lecture by D. E. Barnes.

Permissible Doses. (ICRP)

Occupational exposure

General	300 mrem/week
Gonads	100 mrem/week
Skin	600 mrem/week
Hands and forearms Feet and ankles	1500 mrem/week

Members of public

Living near an installation	500 mrem/year

Derived Levels to Give Doses Within the Above Limits

Laboratory surfaces

(a) Direct irradiation by more energetic beta-emitters (in range 0.5–3.0 MeV): 4×10^{-2} $\mu c/cm^2$.

(b) By resuspension of loose contamination leading inhalation: experimental factor 10^{-7} cm^{-1}.

For alpha-emitters (taking lowest (ICRP) MPC in air which is for Pa^{231} at 10^{-12} $\mu c/cm^3$, and working on 40 hr/week) we get maximum permissible surface contamination as 10^{-5} $\mu c/cm^2$.

For beta-emitters, lowest MPC in air is for Ac^{227}: 2×10^{-11} $\mu c/cm^3$ and other low values are Ra^{228}: 4×10^{-11}; Pu^{241}: 9×10^{-11}; Pb^{210}: 4×10^{-10}; Sr^{90}: 3×10^{-10}.

If we exclude the beta-emitters of high atomic number, treating them as special cases, the general limit is given by Sr^{90} and gives the maximum permissible contamination as 3×10^{-3} $\mu c/cm^2$.

Skin

For ingestion via the hands we assume 5 per cent of contamination remaining on hands after washing might be taken in each day. We can obtain a value for the permissible daily intake by taking the (ICRP) MPC in water and multiplying this by the standard daily intake of water (1.1×10^3 cm^3/day during working hours).

For alpha-emitters lowest value is for Ra^{226} at 4×10^{-7} $\mu c/cm^3$ which leads to permissible contamination of 4.4×10^{-3} μc/hand.

For beta-emitters lowest value is Ra^{228}, 8×10^{-7} $\mu c/cm^3$ leading to 8.8×10^{-3} μc/hand, but if we exclude this the value is set by Sr^{90}, 4×10 $\mu c/cm^3$ leading to 4.4×10 μc/hand.

Irradiation of sensitive layer

Direct by energetic beta-emitters, doses 1.5 rem per 168 hr given by 9×10^{-4} $\mu c/cm^2$, but we must consider non-uniformity and assume contamination is on only 10 per cent of hand (30 cm^2). This gives maximum permissible contamination of hands with beta-emitters: 3×10^{-3} μc/hand; and for skin other than hands: 3.5×10^{-4} $\mu c/cm^2$.

With alpha-emitters, irradiation is after absorption only—assuming exponential distribution from 100 per cent on the surface to 1 per cent at the depth of 2 mm—the permissible dose of 1.5 rem/week would be given by 3×10^{-5} $\mu c/cm^2$ and assuming it is on 10 per cent of area of hands, this gives maximum permissible contamination of hands with alpha-emitters: 9×10^{-4} μc/hand; and for skin other than hands: 1.2×10^{-5} $\mu c/cm^2$.

Techniques of measurement of air contamination. J. LABEYRIE, Commissariat for Atomic Energy, Saclay, France.

The various methods evolved by the French Commissariat for Atomic Energy (Saclay) to measure the contamination of the atmosphere in its various aspects, are described:

Measurement of natural substances, A^{41}, C^{14} and H^3, and fission products present in the form of gas, aerosols or precipitation contaminants; measurement of natural or artificial gases and aerosols in the air of laboratories and, in particular, detection of plutonium.

A description is also given of the apparatus used and a number of typical readings are cited showing the concentrations of these various products.

Techniques de mesure de la contamination de l'air. J. LABEYRIE, Commissariat for Atomic Energy, Saclay, France.

Concentrations maximum admissibles dans l'air (CMA)[1] pour les principaux contaminants radioactifs de l'atmosphère

Tableau 1. *CMA et contamination générale*

Z	Nuclide	Rayonnement* energie max.	CMA en μc/ml (ou c/m^3)	Contamination générale moyenne en France (Saclay) en 1958, exprimée en c/m^3	
				origine naturelle	origine artificielle
1	H^3	B 15 keV	$1 \cdot 10^{-5}$	$6 \cdot 10^{-13}$	$7{,}2 \cdot 10^{-12}$
6	C^{14}	B 150 keV	$1 \cdot 10^{-5}$	$6 \cdot 10^{-13}$	$1 \cdot 10^{-13}$
18	A^{41}	B 1,2 MeV G 1,3 MeV	$5 \cdot 10^{-7}$	0	0
30 à 70	Mélange de produits de fission	A et B de quelques MeV	A $5 \cdot 10^{-12}$ B $1 \cdot 10^{-9}$	0 0	très peu $2 \cdot 10^{-12}$ (sol) 1 à $90 \cdot 10^{-12}$ (10 km)
36	Kr^{85}	B 0,7 MeV	$1 \cdot 10^{-6}$	0	$2 \cdot 10^{-12}$
53	I^{131}	G 0,6 MeV	$6 \cdot 10^{-9}$	0	très peu
38	$Sr^{90} + Y^{90}$	B 2,2 MeV	$2 \cdot 10^{-10}$	0	($\sim 10^{-13}$)
86	Rn^{222}	A 5,5 MeV	ensemble: $1 \cdot 10^{-7}$	Vue moyenne $2 \cdot 10^{-10}$ min. $2 \cdot 10^{-11}$ max. $3 \cdot 10^{-9}$	0
84	RaA (Po^{218})	A 6 MeV			
84	RaC′	A 7,7 MeV			
88	Ra^{226} + 55% produits de filiation	A 4,8 MeV	$8 \cdot 10^{-12}$	0	0
92	U naturel	A	$3 \cdot 10^{-11}$	0	0
94	Pu^{239}	A	$2 \cdot 10^{-12}$	0	0

* A = rayons alpha; B = rayons beta; G = rayons gamma.

1. Les CMA sont extraits du *Journal Officiel des Communautés Européennes*, 2e année No. 11 (20 fév. 1959), Annexe III, pp. 235/59. Ces doses sont inspirées des recommandations ICRP du 1-12-1954.

Tableau 2. Techniques employées pour mesurer les concentrations des gaz radioactifs dans l'atmosphère*

Nuclide	Au niveau de la contamination générale	Au niveau de la CMA
H^3	Enrichissement isotopique ($\times 1000$) puis scintillation liquide	Chbre. ionisation a pré-ionisation 1 CMA → $6 \cdot 10^{-13}$ A/100 l. d'air ou: condenser sous forme d'eau puis scintillateur liquide à-10°-C et PM en anticoincidence
C^{14}	Compteur proportionnel à CO_2 (1 l. CO_2 76 cm Hg → 6 imp/min)	Chbre. ionisation 1 CMA → $6 \cdot 10^{-12}$ A/100 l., ou: scintillateur plastique a grande surface
A^{41}	Pratiquement indécelable dans l'atmosphère normale (loin des réacteurs atomiques)	Chambre différentielle à circulation 1 CMA → $2 \cdot 10^{-13}$ A/100 l., ou: scintillateur NaI, Tl
Kr^{85}	Compteur proportionnel sur Kr commercial. En 1958: 1 l. krypton pur → 4200 imp/min	Chambre ionisation
I^{131}	Filtration sur laine de verre + NO_3 Ag (3,6 kg NO_3 Ag/m^2 retient 98% I^-) puis scintillateur Na I, Tl	Accumulation dans filtre charbon (ou laine verre) + NO_3 Ag, puis scintillateur avec Na I, Tl. 1 CMA → 100 imp/sec par m^3 avec géométrie 2π
Rn^{222}	—Ballon 4 l. avec sulphurede zinc sur parois —Filtration du dépôt actif puis comptage alpha et beta —Chbre. ionisation: $3 \cdot 10^{-9}$ c/m^3 → $1{,}6 \cdot 10^{-13}$ A/100 l.	—Ballons scintillateurs au SZn de 100 cm^3 —Chambre ionisation de 1 l. (1 CMA → $5 \cdot 10^{-14}$ A) —Filtration 100 l. sur 100 cm^3 charbon puis comptage GM des beta de RaC
Tn	Filtration de 21 m^3 puis attendre 5 hr avant la mesure que le dépôt actif du Rn (T = 40 mm) ait disparu	

* Les valeurs des CMA et de la contamination générale sont données au Tableau 1. (*Tableau 3 opposite*)

Production of standardized equipment specially adapted to various methods of radiation control. J. Savouyaud, Radiation Control and Radioactive Engineering Service, Saclay, France, and A. Menoux, Protection Study Office, Saclay, France.

Equipment developed in laboratories for making the measurements required by physicists is often used for highly specialized purposes, calling for delicate adjustment and usually involving complicated maintenance. Such equipment is adequate for protection services provided the number of users is comparatively small.

The rapid expansion and industrialization of research centres has prompted the design of equipment flexible enough to meet all the manifold requirements of protection while offering the greatest possible guarantees with regard to both operation and safety. Compared with the customary type of equipment, such equipment, more closely adapted to specific conditions, should effect substantial savings.

A table is given summarizing the limits of sensitivity required for determining the degree of contamination from the various sources of emission according to the form (solid, liquid, dust) in which they occur. Particulars are also given of the limits to be reached for detecting radioactive gases, together with a summary of the sensitivities required for defining limits of irradiation.

In each of the cases covered by the table, a definition has been given of the most suitable equipment for making the measurements. Where permitted by the standard of performance laid down, a general-purpose appliance has been designed, using specially designed probes for the different protection requirements. In addition, the following are presented:

—a detector for contamination in liquids
—a set of appliances for detecting the activity of aerosol conditions
—a series of ionization chambers, with the appropriate ancillary equipment for measuring irradiation levels
—a specialized apparatus for detecting neutrons.

Tableau 3. Techniques employés pour mesurer les concentrations des divers aérosols radioactifs dans l'atmosphère*

Nuclide	Au niveau de la contamination générale	Au niveau de la CMA
Mélange des produits de fission	Filtration sur papier 100 cm^2 de plusieurs milliers de m^3 d'air puis mesure au GM ou au scintillateur alpha, beta ou gamma. Spectroscopie gamma: possible de doser ainsi Ce, Ru, Zr et Nb. Sr visible seulement après séparation chimique	Filtration sur bande papier 10 cm^2 défilante ou sur filtre électrostatique défilant, avec examen permanent au GM ou au scintillateur beta
Emetteurs de rayons alpha: Ra^{225} Uranium Plutonium Polonium etc.	Pratiquement indécelables dans l'atmosphère normale	—Filtration d'environ 1 m^3 sur filtre papier, examen des alpha avec GM ou scintillateur au SZn. Attendre 48 hr pour que les dérivés du thoron (T = 11 hr) aient disparu —Si l'air du laboratoire est déjà fortement filtré, l'attente peut être beaucoup plus courte (1 hr) —Pour détection instant a née: filtrage fixe sur 100 cm^2, puis comptage alpha et beta en coincidences (témoigne des disintégrations RaC–RaC′ ou ThC–ThC′) puis soustraction des alphas défiliation Rn et Tn après calcul automatique: il reste finalement les alphas à longue période Ra, U, Pu, Po etc.

* Les valeurs des CMA et de la contamination générale sont indiquées sur le Tableau 1.

With more specific reference to radioactive gases, a special probe fitted to one of the above pieces of apparatus enables high energy alpha and beta emitters to be detected. A special integrating device is also provided for measuring total gaseous radioactivity.

It should be noted that all this equipment is designed for standardized fittings, certain components being interchangeable.

Block diagrams and technical specifications for the various types of apparatus will be given.

Wherever possible the problem of control (definition of a threshold or emergency level) has been dissociated from the actual measurement.

For instance, we have made:

—for the control of contamination in solids, a detector set with audio warning signal

—for detecting aerosols, a totalizing filter detector, with a filtered sample depending on the working hours of the personnel.

All this equipment is very strongly built and easy to use. The more specialized equipment for measuring purposes is now only used in particular circumstances.

Simultaneous counting of alpha and beta radiation with one detector and its application for measurement of air contamination. J. LIPPERT, Danish Atomic Energy Commission, Risø, Denmark.

Two methods of obtaining simultaneous alpha and beta counting exist: (1) using a scintillation detector with a "crystal" composed of a plastic scintillator plate for the betas covered with a thin layer of zinc sulphide for the alphas or (2) using a proportional counter. Pulses from the detector are fed to a two-channel analyser. Some work carried out on the proportional counter will be discussed.

Once having the pulses sorted out, it is possible to separate the radon background activity of an air filter sample and thus obtain a better measurement of the air contamination. This is possible by using a coincidence in the radon disintegration chain.

Personnel dose measurement by photographic emulsions. GEORGES SOUDAIN, Photometry and Metrology Laboratories, Saclay, France.

This consists of a photographic dose meter calibrated from 15 mr to 800 r. It has three emulsions of different sensitivities, a scale and a development standard bonded to a cardboard backing.

Optical density is measured by diffused incident light. Doses can be measured visually to within 20 per cent on average. With the help of the developing standard, the dose meter can be treated with a developer having any condition of freshness and temperature.

Session III: *Control of Personnel Contamination.* Chairman: Prof. Dr. H. MUTH (Germany).

Evaluation of body internal contamination using chemical methods. MORRIS F. MILLIGAN, Health Division, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, U.S.A.

Monitoring and air sampling procedures furnish data about the environment in which an individual works and lives; measurement of his internal contamination may be made either directly (by gamma spectrometry or whole body counting), or indirectly (by radiochemical analysis of the urine, blood, faeces, or suitable tissues).

Radiochemical methods should be selected or developed which are specific, sensitive, accurate, and economical. Since it is seldom possible to satisfy all these criteria simultaneously, compromises are usually effected. Methods for the determination of alpha emitters must isolate the desired constituent in a state of high purity, while methods for beta and gamma emitters may be less rigorous.

Results obtained in the laboratory by radiochemical analysis of biological samples refer indirectly to the amount and state of radioactive substances in the body, and are therefore subject to interpretation. Discrimination based on experience must be a part of the interpretation which depends upon the specific nuclide considered and the working history of the individual.

An effective program, in addition to meticulous laboratory techniques, must include frequent sampling of personnel, radiochemical analysis of samples, and interpretation of analytical results.

Critical survey of methods for the determination of radionuclides in urine. F. J. WOODMAN, Chemical Services Department, U.K.A.E.A., Windscale Works, Cumberland, U.K.

Difficulties associated with the analysis of urine for radionuclides are briefly discussed together with some solutions to these problems.

A critical survey of methods is presented with special reference to those in use in the U.K. including procedures for natural and enriched uranium, plutonium, tritium, strontium and caesium.

Performance characteristics and experience in use under routine operational conditions over several years are outlined.

Pooling experience of wounds contaminated with insoluble uranium compounds. H. WIJKER, Health Physics Department, KEMA, Arnhem, Netherlands.

The maximum permissible amount of insoluble radioactive material in cuts or wounds is often so extremely small that it cannot be measured. Therefore, cleaning the wound by removal of some adjacent tissue has become common practice. However, there are cases where this sort of treatment would be better avoided. It is suggested that, after covering the wound for some time with a pad soaked in an absorbing agent, the measurement of the amount of radioactive material on this pad may give some indication whether the wound has to be cut out or not. Therefore it is necessary to know the ratio between the amount of material drawn out and that which remains in the wound. As the cases from which this ratio can be obtained are rather rare, co-operation is suggested in standardizing the treatment and measurements and pooling the results.

The use of low level scintillation spectroscopy in the evaluation of radioactive contamination of the human body. C. E. MILLER and H. A. MAY, Radiological Physics Division, Argonne National Laboratory, Lemont, Illinois, U.S.A.

An instrument which can be used to locate, identify, and measure very small amounts of gamma ray activity (less than 10^{-9} c) within the intact human body has many applications. Three of the most obvious uses are (1) direct measurement of the total body contamination due to industrial exposures without the need of laborious and often misleading excretion measurements, (2) precise determination of biological half-times in the various organs of the body under various conditions of intake, and (3) localization and long-term behaviour of tagged chemical compounds of interest in biological and medical investigations.

To perform these studies, the instrument must always possess high sensitivity and good energy resolution. When measuring the total body activity, it should yield a counting rate which is independent

of body build, and, on the other hand, when measuring the activity in organs, such as the lungs, it must have primarily good position resolution. These two conditions cannot be met with any single detector-subject arrangement, but require the use of two separate techniques.

The various factors which limit the sensitivity obtainable with gamma ray spectrometers have been studied in detail. The gamma ray background of the shielded volume has been systematically analyzed and steps have been taken to reduce the background to a minimum. This has required that all materials of construction be measured and chosen on the basis of lowest specific activity.

In addition, the possibility of eliminating the effects of cosmic rays has been studied using anti-coincidence techniques and by making measurements 163 m.w.e. underground.

The various crystal-to-body geometrical arrangements which may be used to measure the total body activity have been evaluated. Two arrangements, a one-meter-arc technique and a tilting chair position, yield the most satisfactory results.

Several spectral techniques have been investigated and will be described. One is a two crystal summing-coincidence arrangement for measuring small quantities of radium or thorium in the presence of large quantities of other radioactive elements. A second special technique was designed to measure the Ra^{226}/RaB partition ratio directly in the living intact human.

The application of gamma-ray spectrometry in Health Physics other than for body monitoring. D. H. PEIRSON, Health Physics Division, A.E.R.E., Harwell, Berkshire, U.K.

1. *Introduction*

The rapid analysis of radioactivity by identification of the characteristic gamma-ray spectra from radionuclides has made gamma-ray spectrometry a powerful technique of measurement in Health Physics. The technique is first illustrated by spectra of general interest. Examples of particular interest to the Health Physicist are presented below.

2. *Occupational contamination*

Examples of spectra together with case histories are given of contamination from reactor fuel rods and graphite in an engineering workshop, in an air sample from a remote handling cell, and in urine.

3. *Nuclear weapon debris*

Concentrations of fall-out are monitored at various altitudes in the atmosphere and in rain. A spectrum of very low activity is shown together with the variation with time of various fission products in rain.

4. *Environmental survey of biological samples*

The gamma-ray spectrometer has been used to study the uptake of certain fission products from soil into plants. After the accident at Windscale it proved invaluable in routine assay of milk, grass, vegetables, etc. for I^{131} in the presence of other fission products.

Examples of the practical use of a gamma spectrometer in the detection of radioactive contamination of the human body. L. JEANMAIRE, Radiotoxicology Laboratory, Commissariat for Atomic Energy, Saclay, France.

The apparatus used by the Commissariat for Atomic Energy contains a sodium iodide crystal 20 cm dia. × 10 cm high, a linear amplifier and a 25-channel selector. Protection, which is relatively light for an assembly of this type, is obtained simply by a 5 cm thickness of lead.

This spectrometer is sensitive enough to register the natural radioactivity of the human body. It will also quickly detect accidental contamination by a gamma emitter, even in doses which are considerably smaller than the maximum permissible doses recommended by the ICRP.

The apparatus can also be used for taking measurements after neutron irradiation.

Despite certain difficulties, this system for detecting internal contamination can serve a very useful purpose in current practice.

Session IV: *Practical Problems of Health Physics in Specific Types of Installations.* Chairman: Mr. G. JENSSEN (Norway).

Confinement techniques and handling of plutonium in research laboratories. A. R. KEENE, Hanford Laboratories Operation, General Electric Company, Richland, Washington, U.S.A.

Plutonium, with its extremely low maximum permissible body burden, requires that some degree of confinement be provided in order to handle the material safely in research laboratories. Dependent upon the quantity and physical form of the material being handled, confinement techniques may range from complete physical isolation of the material from the research worker to nominal control through the use of standard survey procedures. Confinement and handling methods must consider the plutonium both as a source of external radiation and as a bone-seeker, potentially most likely to enter the body

through accidental inhalation or skin injury. Other factors to be considered in establishing safe handling procedures include pyrophoricity of plutonium metal, required ventilation, use of strippable coatings, and survey techniques. A review of problems likely to be encountered when handling plutonium in research laboratories is made.

A method for scintillation counting of very low energy particles (below 1 keV). M. FORTE and A. ANZANI, Health Physics Division, C.N.R.N. Centre of Ispra, Italy.

A method is described for analysis of time sequence of cathode photo-electrons during the scintillation of very low energy particles. It is actually possible to discriminate, from single dark cathode electrons, scintillation pulses of 2 or more grouped photo-electrons. With high efficiency scintillators (e.g. ZnS*), particle energy requested for a photo-electron pair is below 1 keV. Particles of this energy are discriminated with high efficiency. This method, which avoids photo-multipliers cooling, is proposed for tritium counting.

Recent progress in the design and operation of high activity laboratories. R. BAZIRE, Section for Protection against Radiation, Saclay, France, and F. DUHAMEL, Radiation Control and Radioactive Engineering Service, Saclay, France.

1. *General principles*

The design of high activity laboratories depends closely on the methods used for guarding against external exposure and contamination.

For this reason the Commissariat for Atomic Energy established its "Radioactive Engineering Service". As, however, capital outlay remains one of the chief considerations, the design must always be as simple as possible and this means that radiation controls must be both flexible and constructive.

By linking together radiation control and radioactive engineering, a single theoretical and practical approach to design and procedure becomes possible.

In this way it has been possible to establish:

—practical exposure standards
—systems of graduated confinement, according to the degree of radioactivity
—mapped out areas with regard to traffic-flow (ventilation, movements of staff or equipment)
—methods for the evacuation of radioactive waste.

These items do not exhaust the list of considerations and a wide range of other possible solutions still remains.

2. *Examples of schemes devised or applied in France*

Various designs for high activity laboratories built in France are examined:

—*The Saclay High Activity Laboratories* (the oldest); permissible limit 100 c
—*Laboratory for Examining Irradiated Fuels* (prototype based on experience in the United States); permissible limit 10,000 c
—*Radioisotope production laboratory* (under construction) to an original design: work is done in a cold zone, easily isolated
—*Very High Activity Laboratory of Grenoble* (planning stage) a replica of the Saclay L.E.I.F., adapted to suit requirements at Grenoble
—*Alpha, beta, gamma Laboratory of Châtillon* (low-cost scheme, using special equipment, with combined laboratory and industrial workshops)
—*Caissons* (system whereby an industrial shop can be converted to a high activity laboratory for dealing exclusively with alpha radiation)
—*Large size alpha cells* (system for high productive capacity)
—*Special case: Hot cell for pile* (clearly defined specialized operation in a plant which cannot be otherwise adapted).

3. *Protective materials*

Questions now under study in France, including some which have become quite familiar in the United States and the United Kingdom (concerning concrete, special glass, paint and cladding materials, tanks for effluents, radiation-resistant materials) are reviewed.

4. *Mechanical equipment*

The handling devices used in France are listed, and a brief account given of those manufactured in that country, together with miscellaneous equipment developed for high activity laboratories (periscope, dust removal device with recycling, movable sink, container for decanting highly active liquids).

5. *Operational methods*

Design and operation go hand in hand. Substantial economies in building costs can be achieved by sound operational planning, without detracting from safety.

Control is decentralized but the warning system is centralized and safety measures are applied practically instantaneously before a high dose of exposure is reached. The wide spread use of polyvinyl sheets facilitates operations, and spontaneous precautions taken by those handling equipment helps to reduce hazards.

Continuous sampling devices enable a constant check to be kept on the proper working of the plant.

Licensed collecting service for radioactive waste. H. R. MARCUSE, Radiological Protection Section of the Health Council, The Hague, Netherlands.

Since the principle of containment with respect to ionizing radiation and radioactive substances is generally accepted, and the number of users of these agents is increasing in hospitals, laboratories and industry, one of the consequences in the Netherlands is the establishment of a licensed collecting service for radioactive waste.

An evaluation is made of the kind and the quantities of waste that must be collected in the near future.

An analysis will be given of the technical problems expected and the facilities needed for maintaining public health and safety.

Session V: *Practical Problems of Health Physics in Specific Types of Installations (continued)*. Chairman: Mr. H. WIJKER (Netherlands).

Progress and experience in decontamination techniques. J. NEIL, Radiation Hazards Control Branch, Biology and Health Physics Division, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada.

An outline of progress in decontamination techniques, based on experience at the Chalk River plant of the Atomic Energy of Canada Limited, will be presented. The following will be discussed:

—Early methods of contamination control and methods of decontamination
—Advantages of centralization
—Early methods of personnel protection against contamination, including respirator and protective clothing
—Organizing and planning of decontamination work following a major reactor accident
—Present decontamination methods including the use of an automatic dip washer, an ultrasonic cleaning device, steam jets, solvents and other cleaning agents
—Decontamination installations including a building for small equipment and one for vehicles and larger equipment
—Outline of plans for future facilities and equipment including a building for the decontamination of articles up to fifty tons, a mobile change room, a mobile decontamination trailer, a remote monitoring device, portable shielding, shielded vacuum cleaners and automatic wall and floor cleaners.

Radiological problems in operation of small research reactors. W. JACOBI, Nuclear Chemistry Section, Institute for Nuclear Research, Berlin-Wannsee, Germany.

The neutron flux in the centre of the core of most small research reactors with a power of 10 kW–1 MW is in the order of magnitude 10^{11}–10^{13} n/cm² sec. The saturation activity of fission products built up in the core is within the range of 50 kc–5 Mc. The number of experimental facilities which can be used at the same time is small because of the high change of reactivity caused by input probes. Therefore experimental work at the reactor changes more frequently than with higher power research reactors and good programming is necessary.

Problems of organization arise from the fact that these reactors are operated in most cases by university institutes for study, training and research. A permanent group of personnel is essential for the safe operation and control of the reactor and for health physics work. Such an organization is sometimes unusual for small institutes.

Size and control of radiation level in the reactor hall during loading and unloading the reactor, experiments at open channels during normal operation as well as the necessary monitoring equipment for gamma- and n-radiation and air contamination, are discussed. Problems of corrosion in the reactor core and tank, and fuel element leakage, leading to contamination of primary cooling water, are also described.

Finally questions concerning reactor-safety and hazard-analysis in connection with the conditions at the Berlin research reactor dealt with and conclusions drawn as to the construction, tightness, ventilation system and airlocks of the reactor hall.

Air pollution observed at the Swimming Pool Reactor SAPHIR. R. BERNER, W. A. HUNZINGER and T. HÜRLIMANN, Reaktor A.G., Würenlingen, Switzerland.

After one year of running the Swimming Pool type reactor SAPHIR a sudden rise occurred in the air pollution by a factor of nearly five. During the same time the concentration of fission products in the pool water did not change. The cause of the air pollution was due to a change in the cooling water circulation.

Analysing air samples by gamma-spectroscopy and

decay rates after chemical separation the following isotopes were identified: Cs^{138}, Rb^{88}, Rb^{89} and Sr^{89}. No halogens were found. The separation of Cs and Rb was carried out by ion exchange on ammonium-molybdatophosphate, $(NH_4)_3[P(Mo_3O_{10})_4]$.

After a few hours running at 1 MW, concentrations of 10^{-9} μc/ml for Cs^{138} and 10^{-8} μc/ml for both Rb isotopes were observed.

Attempts are made to explain quantitatively the air pollution from the known amount of fission products in the pool water and to estimate an upper limit for all fission products in the air, descending from noble gases.

Radiological and organizational problems in the control of criticality. B. A. J. LISTER, Health Physics Division, A.E.R.E., Harwell, Berkshire, U.K.

An analysis of the accident history of the Atomic Energy industry indicates that the type of accident most likely to have serious consequences is that resulting from the attainment of an unwanted condition of criticality. The conditions normally encountered in laboratories probably completely exclude the possibility of a violent atomic explosion and the accidents so far experienced have been accompanied by little physical damage to the installation.

However, the accompanying radiation can constitute a serious hazard to people in the vicinity and lethal doses could result with little outward sign that an accident had occurred, unless special instruments and alarms had been provided.

All the criticality accidents so far recorded have produced a total number of fissions in the range 10^{16}–10^{18} (equivalent to an energy release of 0.1 to 10 kWh). Whereas in most cases the energy has been released in only one burst, in another a number of pulses have occurred over a period of some minutes.

This paper refers specifically to the way in which criticality control can be handled in a research establishment and the two complementary aspects of accident prevention, knowledge and appreciation of the knowledge, are considered in turn.

In the last few years many laboratories have contributed a considerable amount of background research in order to provide a basic understanding of the relevant nuclear and neutron physics and the development of mathematical methods of predicting nuclear behaviour. Useful reviews of the subject have been published. Although it is possible to build up a certain "feel" for the subject, and although this "feel" may often lead to satisfactory solutions for many problems that occur in experimental establishments, it is not good enough to rely for safety on initiative and it is necessary to call on the help of mathematical and nuclear physicists backed by experimentalists.

Having ensured an adequate level of knowledge, training and criticality consciousness throughout an establishment, it is necessary to make certain that only safe practices are carried out. In order to minimize chances of accidents resulting from human fallibility, certain managerial and administration procedures have generally been adopted. These include:

(a) definition of responsibility
(b) insistence on the proper thought and preparation in each case of possible accident, usually by demanding a written application and safety statement by the responsible officer
(c) subjection of each application to independent expert criticism
(d) the appointment of an independent criticality inspector to check that safety conditions are observed.

The responsibility of the most senior person directly concerned with any activity for the safety of this activity cannot be avoided. Safety services must be considered advisory. The preparation of safety documents and the perusal of applications for any use of fissile material by a committee of experts is a time consuming operation but it is considered of primary importance.

It is frequently possible to design equipment for experiments involving fissile material in such a way that the loss of neutrons from the system is so great that accidental criticality is impossible. It is not, however, the intention in this paper to discuss the information on which such designs can be based.

In experimental laboratories it is not always possible or expedient to use ever-safe equipment and in such cases it is necessary to employ material control methods. Such methods impose severe management problems, particularly in a research establishment where quite considerable quantities of potentially dangerous materials may move from laboratory to laboratory. In the final analysis safety depends on discipline and adequate methods of accounting and of control of movements of material.

Although the measures already described should go far to avoid the occurrence of accidental criticality, plans must be laid for action in the event of such an accident happening. The need for a reliable warning system is of prime importance. The first few seconds after a critical condition has been produced are vital and experience has shown that the rate of initiation of an audible warning after an increase in radiation and the speed of reaction of personnel are paramount factors. The latter emphasizes the need for training and indoctrination of staff and the importance of

frequent exercises. Evacuation is a factor which must be considered at the design stage.

After an accident has occurred there is the need for reliable communications and the establishment of control centres is a point which must not be omitted from preliminary planning.

Necessary action after an incident of this kind may well be hampered by the number of persons who may be involved, even though perhaps not directly concerned with the equipment responsible for it.

It is clearly essential to have early information on the dose received and to be able to find and segregate those who have received high doses of radiation in order to enable medical practice to be put into effect. It is also necessary, but at a later date, to have information on the true biological dose for research purposes.

It is not the purpose of this paper to discuss methods of dosimetry, nor the treatment of cases of over-exposure. It is anticipated that these subjects will be covered by later papers.

Some problems in the control of criticality. K. Z. MORGAN, Health Physics Division, Oak Ridge National Laboratory,* Oak Ridge, Tennessee, U.S.A.

Following the criticality accident at the Y-12 Plant in Oak Ridge on June 16, 1958, a Radiation Emergencies Steering Committee was formed. This Committee consisted of representatives from the X-10, Y-12 and K-25 plants in Oak Ridge, Tennessee, and the gaseous diffusion plants at Paducah, Kentucky, and at Portsmouth, Ohio. This Committee made a comprehensive review of the various problems that must be considered in the prevention and handling of criticality accidents, and the principal purpose of my few remarks will be to review and comment on some of the salient features of this study by the representatives from these five organizations.

Nuclear safety depends upon *design* and/or on *administrative control* with a minimal dependence on administrative control—a dependence commensurate with economic practicability. In any case nuclear safety must be based primarily upon the results of criticality experiments and when experimental data is not available, calculations may be used if the limits of error are stated and appropriate safety factors are applied.

Perhaps administrative control should be considered to be economically practicable primarily for short-term projects, while for long-term projects a continuous train of safe processing equipment should be designed and set up where feasible. Almost without exception, administrative control should require the occurrence of at least two independent contingencies for a criticality accident to be possible. As a general rule, a double or multiple-contingency requirement should be met by all *safe* equipment over which administrative controls are exercised.

Most of the design control factors relate to *geometric restrictions*. However, where adequate experimental data confirm the mechanical and/or chemical stability of *fixed neutron absorbers* and the nuclear safety of process equipment incorporating them has been established experimentally, such equipment is regarded as "safe". The use of Cd $(NO_3)_2$ or other liquid nuclear poisons in the cooling coils of equipment handling highly enriched uranium or plutonium offers some protection in most cases; however, the use of such nuclear poisons must be undertaken only after a careful analysis of each application since they may actually increase the hazards of criticality.

One of the situations considered most likely to lead to difficult criticality control is that in which areas under design control meet or overlap areas under administrative control. This dangerous change or possible overlap in philosophy of criticality control should be avoided unless there is no practical alternative. Such a situation set the stage for the Y-12 accident on June 16, 1958, when enriched uranyl solutions passed from the B-1 area that was under design control into the C-1 area that was under administrative control and where *unsafe* containers were not forbidden.

An important part of criticality accident prevention depends upon an adequate review procedure. An accurate inventory of all fissionable material must be maintained at all times and there must be a constant vigilance and awareness of the problems of nuclear safety on the part of personnel at all levels of supervision. Effective means of communication must be maintained especially when there is a transfer of responsibility resulting from the passage of materials from one area to another or occasioned by personnel changes, e.g., changes during lunch hour. In general, such communications should be written as well as explained orally. Education programs in nuclear safety should be conducted repeatedly among personnel at all levels in the organization so that each employee understands his responsibilities at all times and is aware of how he can contribute most to nuclear safety not only during day-to-day routine operations, but also during emergency situations.

In the final analysis primary dependence must be placed in criticality measurements and calculations because there have been so few criticality accidents that furnish useful guidance, and in such cases we are limited primarily to studies of dangerous situations

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

that were discovered and corrected. In general the more basic criticality measurements cannot be made in situ but must be conducted in especially constructed criticality facilities. The primary effort should be directed toward obtaining the more fundamental experimental and theoretical criticality data of the widest application, but many engineering experiments should be conducted rather than rely heavily on extrapolations and interpretations. Criticality codes used on high speed computing machines not only save an immense amount of time but make it practical to check a great variety of combinations of the parameters.

Framing of practical rules for safety from contamination. F. DUHAMEL, Radiation Control and Radioactive Engineering Service, Saclay, France, and J. LAVIE, Radiation Control and Radioactive Engineering Service, Saclay, France.

On the basis of the general standards recommended by the ICRP, rules for the avoidance of irradiation risks can be worked out fairly easily, but contamination risks are more complicated to deal with.

One of the fundamentals for framing rules of this kind is a proper classification of the substances to be handled. Although many classifications have been published, they are not entirely satisfactory.

The classification suggested is based on the concept of a maximum inhalable quantity. It is found convenient in practice to take into account the product of the maximum permissible concentrations in the air, expressed successively in terms of c/m^3 and g/m^3.

This method can then be used to draw up a double chart on which equal risk correlations can be read on a diagonal.

The various recognized classifications are shown on the same chart for purposes of comparison.

By taking into account a factor to cover the physical condition of the substance and another factor representing the type of operation, the risk of a given operation with a given radioactive substance can be measured in terms of quantity.

In the light of the figures thus obtained, rules can be drafted for the handling, storage or transport of radioactive substances. The special case of fissionable materials necessarily calls for separate treatment.

Session VI: *Techniques of Radiation Measurement and Shielding Problems in Relation to High Energy Accelerators.* Chairman: Mr. F. ALDER (Switzerland).

Radiation security and control at the Saclay proton synchrotron. H. JOFFRE, P. CANDES and A. STIRLING, Protection Control and Supervision of Piles and Accelerators, Saclay, France.

The performance and special characteristics of a proton synchrotron involve unusual problems of protective shielding and detection of radiation.

Particulars will be given of the exposure levels reached at various points in the building under routine conditions at the Saclay synchrotron, and of the modifications proposed with a view to enhancing the efficiency of the present shielding.

In monitoring fluctuations in levels of gamma radiation and fast neutrons around the accelerator, new types of detectors have had to be evolved to deal with the pulsating flux of high energy particles and gamma radiation. The system of measurement adopted consists of a repetition cycle in the course of which the following operations are effected in turn: integration of ionization current or pulses with a charge capacity for about 3 min, measurement and recording of the value of the charge, and discharge of capacity. This method is used for measurements made in ionization chambers and scintillation detectors, both for fixed detectors and for some kinds of portable inspection detectors.

Other methods of measurement are also used in order to get a better knowledge of the energy spectrum of the radiation emitted: threshold detectors, coincidence telescope, Cerenkov counter etc.

To provide for the safety of personnel responsible for operating or using the accelerator, an elaborate warning and security system has been installed. It includes:

(a) local arrangements:
—a system for indicating the working of the accelerator and level of radiation
—safety locks for securing entrances
(b) a central control system for indicating all readings, signals and checks on a radiation control panel.

A round-the-clock shift system ensures proper supervision of experiments and of the radiation control panel.

Survey techniques for accelerators. B. M. WHEATLEY, CERN, Geneva, Switzerland.

Survey techniques in use on the 600 MeV synchro-cyclotron and the 50 MeV linear accelerator at CERN are briefly described, together with proposed methods for surveying the 25 GeV proton synchrotron. Methods of analysing the radiation field are discussed, with special reference to ion chamber techniques for separating the gamma and heavy particle contributions. Proportional counting and activation techniques for investigating the heavy particle spectrum are considered and suitable apparatus is described.

The shielding of high energy accelerators. L. R. SOLON, Health and Safety Laboratory, U.S. A.E.C., New York, N.Y., U.S.A.

The construction of particle accelerators of ever increasing energy is continuing on an international scale at an unprecedented pace. These devices are enormously expensive, both in terms of capital cost and in terms of the precious reservoir of scientific and engineering manpower that each of these machines requires for adequate utilization.

For full exploitation of the particle accelerator, it is essential that the machine and its associated facility be suitably shielded. The consequences of insufficient shielding can result in important limitations of beam current or allowable experimental configurations.

Though one often thinks of these limitations in terms of not exposing research personnel to inordinately high doses of ionizing radiation, there is another limitation which is at least equally important for most contemporary accelerators. For many experiments in high energy particle physics, particularly those involving the more exotic nuclear detectors such as the bubble chamber, a low level radiation environment is necessary.

The required radiation level is, in fact, frequently below the level which is acceptable to personnel in terms of safety. Those of us in the radiation protection business should not regard ourselves as on a crusade to protect accelerator researchers against their own folly. Radiation shielding for the protection of personnel and radiation shielding for the performance of good experiments are two sides of the same coin.

In this discussion, we shall deal principally but not exclusively with the problems associated with the shielding of very high energy machines. Such a machine shall be defined, somewhat arbitrarily, as one capable of producing mesons in reasonable yield—implying that the energy of the primary beam be 200 MeV or greater. By concentrating attention on such devices we do not intend to suggest that shielding problems for lower energy accelerators are thoroughly resolved. However, the fact of the matter is that these lower energy machines have been built in such numbers that abundant practical shielding information is available, at the very least, on a purely empirical basis.

The situation with respect to meson producing machines, particularly those built or under construction in the Bev region, is not nearly so salutary.

Here the parameters necessary for good shielding evaluation are generally not known accurately. The cross-sections, yields of secondary particles, scattering data and other types of fundamental information which would permit a genuinely quantitative shield design are in fact not available and cannot really be known until the accelerator has been operating for a substantial period of time.

Thus the shielding design depends upon theoretical work and venturesome extrapolation of data obtained at considerably lower energies. Accelerator shielding can almost be defined as a combination of skilful conjecture and engineering judgment to guard against the Scylla of excessive stray radiation and the Charybdis of bankruptcy which haunts all these enterprises.

It is the purpose of these remarks to set forth in a more or less detailed way the approaches that have been taken with respect to shielding design at a representative group of United States accelerators, what American experience has been, and what areas this observer regards as presenting the problems of greatest urgency. Though we shall not shy away from arithmetical formulations where these may be useful, it should be stated in advance that numerical values are generally only intelligent estimates and should be treated with appropriate caution.

For this presentation, the author shall rely very heavily upon the results of the "Conference on Shielding of High-Energy Accelerators" held in New York in April, 1957 under the auspices of the U.S. Atomic Energy Commission Health and Safety Laboratory and Division of Research.

He shall wherever possible make the appropriate attribution for the ideas which he reports. Because so many of the concepts were evolved in spirited panel discussions during the conference, it will not always be possible to identify the contributor.

The application of a biological dosemeter. B. LARSSON, The Gustaf Werner Institute for Nuclear Chemistry, Uppsala, Sweden.

It is a well-known fact that two absorbed radiation doses, although energetically equal, do not always give the same effect in a given biological system. One of the most important factors is the linear energy transfer (LET) along the track of the ionizing particles. The relative biological efficiency (RBE) of radiation on tissues, cells and cell structures generally increases with increasing LET. In order to estimate the biological hazards of a certain irradiation procedure—the principal problem in health physics—it is possible to estimate the contribution to the absorbed dose for all parts of the energy spectrum of each radiation component and then to perform an integration with assumed values for the varying RBE. Even if always physically possible, this procedure is often complicated, and the result may

be very approximate because of lack of knowledge of the various RBE-factors involved. Another possibility is to use as dose indicator a biological system in which the radiation effects to be analysed respond to variations in LET in approximately the same way as most other radiation effects in animals.

In an investigation performed with the synchrocyclotron at Uppsala, scoring of chromosome aberrations in irradiated broad bean seedlings was used to get quantitative information on the biological efficiency of the mixed secondary radiation from materials irradiated with high energy protons. The test system was easily calibrated in rem/hr by means of X-rays.

"Biological dosemeters" of the type described are reliable, tissue-equivalent and fairly radiosensitive and might find some further application in health physics at high energy accelerators.

Session VII: *Emergency Situations*. Chairman: Dr. C. POLVANI (*Italy*).

Immediate evaluation of external radiation doses and internal contaminations in the event of a radiation accident. K. Z. MORGAN, Oak Ridge National Laboratory,* Oak Ridge, Tennessee, U.S.A.

There are two types of radiation exposure during accidents: (1) exposure to external sources and (2) exposure from internal sources. The first and most important consideration in radiation accidents is the prevention of such accidents. Appropriate training programs, the proper design and operation of buildings and facilities, and the effective functioning of an adequate health physics program cannot prevent all radiation accidents, but they can reduce the probability of their occurrence and can minimize their serious consequences. The second important consideration is early warning and immediate action when an accident happens. There is no doubt that the prompt exit of personnel during the Y-12 accident on June 16, 1958, and during the Argonne accident on June 2, 1952, prevented more serious consequences; and had there been similar retreat from the scene of the first two serious Los Alamos accidents (August 21, 1945, and May 21, 1946), there is reason to believe the human exposures would have been materially reduced, and perhaps the two fatalities could have been prevented. An important part of the prompt action during the period of a radiation accident is the immediate identification of the individuals who received high exposures. Several features of personnel monitoring meters enable one to make this identification. The third step in an accident is a careful estimate of the absorbed dose from the various types of radiation, i.e., from neutrons, gamma radiation, beta rays, etc., received by each of the accident victims. In addition, information should be obtained on the energy spectra of the radiation, the dose distribution throughout the body, and the dose rate. All these factors furnish information that is useful in determining the seriousness of the exposure and in deciding on the subsequent medical treatment. The absorbed dose information is obtained from the personnel monitoring meters, from area meters, from the activation of Na^{24} and P^{32} in the body, and from a study of the radiation source and the flight path of each of the exposure victims. The fourth and last step in a radiation accident is that of re-entry and cleanup. Sometimes these steps must be taken quickly but with considerable caution and forethought. For example, a critical assembly that resulted in the accident may go critical again with the slightest provocation, or in the case of a radium spill, cutting off or isolating part of the ventilation system may prevent serious human exposure and contamination resulting in a great financial loss.

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

Recent progress in internal decontamination methods. A. CATSCH, Institute of Radiobiology, Reactor-Station, Karlsruhe, Germany.

Up to now ethylenediaminetetraacetic acid, a chelating compound, has frequently been used as therapeutic or diagnostic agent in cases of poisoning by radioactive metals. It is of very poor efficiency when treatment is delayed and also inefficient in preventing the deposition of certain radio-elements in the skeleton.

Experimental data are presented showing a much higher efficiency for the following polyamino-acids: diaminodiethylethertetraacetic acid and diethylenetriaminepentaacetic acid. Removal of internally deposited radiocerium, radioyttrium, lead and uranium becomes possible, even if the agents are administered *per os* or delayed. So far all attempts to find better means of lowering the body burden of radiostrontium have failed.

Emergency measures and treatment of accidental over-exposure to radiation or radioactive contamination. H. JAMMET, Health Physics and Radiopathology Service Commissariat for Atomic Energy, Saclay, France.

The primary concern of doctors who have to give emergency treatment to people exposed to external

irradiation or substantial accidental radioactive contamination is to estimate as accurately as possible the exact dose absorbed, or the amount of contaminating radioactive element involved. Only if these things are available is it possible to give correct therapeutic indications. However, in view of the uncertainty and difficulty of certain measurements, and above all in the absence of accurate physical data, a number of clinical symptoms may be regarded as essential alarm signals for assessing the degree of exposure.

Cases of accidental over-exposure may be differentiated according to whether the subject has been exposed to partial or total irradiation. In the case of acute exposure to total irradiation by several hundred rem, urgent initial shock treatment is indicated, as specific treatments for restoring the organs affected, or temporarily taking over their function, should only be applied later in the light of clinical developments. Should the dose exceed 1000 rem, drastic action must be taken from the start, without waiting to see what course the clinical symptoms will take.

In cases of serious partial exposure, in particular to radiation of low penetrating power, the principal area for medical attention is likely to be the skin. Owing to the difficulty of assessing the actual dose received, it is necessary to await clinical developments. In any event it is recommended that delay action be taken to limit trophic or infectious complications of the patient's lesions. The next stage will be restorative treatment, the indications for which must be carefully pondered.

In cases of accidental radioactive contamination it becomes necessary to distinguish between superficial or cutaneous contamination and deep contamination accompanied by the incorporation of radio-elements. In either eventuality, the first concern must be to prevent by all means available the radioactive contamination from spreading and then to decontaminate the organism. Cutaneous decontamination is a comparatively simple matter, using the most ordinary skin-cleaning methods, and only occasionally calling for anything more drastic, in which case care must be taken to avoid irritating or injuring the natural protective layers.

In cases of transcutaneous internal contamination (wounds) through the respiratory or digestive systems, the first concern is to reduce the absorption of radio-elements through the system affected, by cleansing of the wound and the administration of medicaments capable of converting the radioactive substances to a form in which they cannot be assimilated by digestion. This may be supplemented by any treatment likely to eliminate as much as possible of the radioactive intake before it reaches the internal organs (expectorants, enemas, purges etc.). The next stage must be to eliminate from the internal regions, and in particular from the organs of assimilation, any contaminating radioactive substances. In most cases these forms of treatment are still extremely disappointing, relying as they do on chelation or complex compounds. Finally, at a third stage complaints arising from irradiation of the body's organs through the introduction of radioactive substances into the system may have to be treated; in such cases treatment is symptomatological.

Some lessons from the Windscale accident.
B. S. Smith, Health Physics Division, A.E.R.E., Harwell, Berkshire, U.K.

As a result of the accident to the Windscale Pile in which parts of the graphite and some of the fuel were over-heated, some 20,000 c of 8 day I^{131} were discharged up the stack. The release occurred over several hours during which the meteorological conditions changed, but the main pattern of deposition was produced in a south east direction. The magnitude of the resulting deposition and its variation along the axis of the plume is compared with that predicted by Chamberlain and Megaw in their calculations of the hazards resulting from reactor accidents.

Some forecasts are made of the exposures which might have been received by personnel at short distances, had the discharge occurred at a level very much lower than the top of a 450 ft stack.

The observed relation between iodine on grass, the corresponding gamma radiation at 1 m and the resulting level of contamination in milk is given. The problem of delineating by gamma monitoring the limits of grass contamination which will give rise to the maximum U.K. emergency tolerance in milk (0.065 μc I^{131}/l.) is discussed.

Closing Remarks by Dr. K. Z. Morgan

On behalf of the U.S. delegation and all the Health Physicists assembled here from many countries I would like to thank the Danish AEC, the European Nuclear Energy Agency of the Organization for European Economic Cooperation, and all the other organizations that have made this Conference possible. In order to convey our appreciation and to perpetuate the significance of this first such Health Physics

Conference in Europe, we would like to present to Risø an oak tree; the first radiation protection instrument which offered protection from the sun's radiation. May it grow and flourish and for many generations signify the importance of Health Physics. This package I am presenting on behalf of all Health Physicists was procured by Professor SIEVERT and contains not one but four oak trees. This of course is to increase the probability that one may survive in spite of the uncertainties of the forces of nature. I now call on Professor SIEVERT, one of the first and most distinguished Health Physicists, to make a few closing remarks.

Closing Remarks by Prof. Dr. R. M. SIEVERT

Ladies and Gentlemen.

Will you permit me to express to the Organizers of the Risø Symposium special thanks from the Scandinavian participants—from your brother countries. It might be appropriate that these thanks are submitted by a representative of the coming generation, the generation which we all hope will be able to see the results of the present efforts in the nuclear energy field. They will, I hope, one day understand how much we are indebted to the new kind of work which is going on in this field, based on co-operation between outstanding administration and deep scientific knowledge. This little girl of two and a half years, who brings you these flowers, is a real Scandinavian girl, as her father's mother was born in Oslo, her mother in Copenhagen and her father in Stockholm.

A LIST OF PAPERS FOR PUBLICATION IN FUTURE ISSUES

R. GOLDEN and E. TOCHILIN: Characteristic curves from different ionizing radiations and their significance in photographic dosimetry.

R. GARRETT and J. S. LAUGHLIN: A diagnostic X-ray exposure dose chamber.

A. L. BONI: Rapid determination of mixed beta-gamma radionuclides in urine.

H. L. BUTLER and R. W. VAN WYCK: Integrity of vinyl plastic suits in tritium atmospheres.

E. C. WATSON: A film technique for measuring the exposure dose from plutonium.

C. W. SILL and J. K. FLYGARE, JR.: Iodine monitoring at the National Reactor Testing Station.

WRIGHT H. LANGHAM: Physiology and toxicology of plutonium-239 and its industrial medical control.

JAY BOYD BEST: Maximum likelihood analysis of ionizing radiation induced mortality in whole body fractionated dose experiments.

JAY BOYD BEST: Theoretical relationship between percent kill and shortened life span measures of radiation injury.

G. S. HURST, R. H. RITCHIE and L. C. EMERSON: Accidental radiation excursion at the Oak Ridge Y-12 Plant—III. Determination of radiation doses.

G. A. ANDREWS, B. W. SITTERSON, A. L. KRETCHMAR and M. BRUCER: The accidental radiation excursion in the Oak Ridge Y-12 Plant—IV. Preliminary report on clinical and laboratory effects in the irradiated employees.

JOHN S. HANDLOSER: Stray radiation dosimetry around pulsed accelerators.

R. H. GOEKE and C. L. WEAVER: Health physics field operations in Atomic Energy Commission weapons testing program.

A. L. BONI: Urinalysis method for enriched uranium.

R. L. BLANCHARD, B. KAHN and R. D. BIRKHOFF: The preparation of thin, uniform, radioactive sources by surface adsorption and electrodeposition.

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ACCIDENTAL RADIATION EXCURSION AT THE OAK RIDGE Y-12 PLANT—III

DETERMINATION OF RADIATION DOSES

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I. INTRODUCTION

An analysis of the physical events leading to an accidental radiation excursion at the Oak Ridge Y-12 Plant has been presented in the first of this series of articles.[1] A companion article[2] is devoted to health physics problems arising from this accident. The present article describes the methods which were used to arrive at the doses received by the individuals. Although our main objective is to describe the dose analysis of this particular excursion, efforts will be made to present methods and information which may be of use in similar problems.

Very early estimates of the doses received by the individuals, based on their known positions at the initiation of the excursion and an estimate of the total number of fissions in the assembly, were unreasonably high. For example, the fast neutron component of the total dose received by employee *A* was estimated to have been 1500 rads. Hence one must assume that the location of personnel during a substantial fraction of their exposure time is unknown. This fact, together with the fact that the exposed employees were not wearing any type of personnel monitoring equipment, dictates that the evaluation of dose be based on the activation of body elements. Experience at Los Alamos[3] in the Pajarito accident indicates that of the body fluid methods, Na^{24} in blood may be the most reproducible criterion of neutron exposure and, in particular, indicates that the formation of P^{32} in urine is poorly understood.

In spite of the fact that the exposed individuals were wearing no dose-measuring devices,† it is believed that the final dose values are reasonably accurate ($\pm$20 per cent). The doses were arrived at by the following procedure: (1) Blood and urine samples were collected from the exposed employees; these samples were counted for radioactive substances‡ (Na^{24} in the blood and P^{32} and Na^{24} in the urine) as described in Section V. (2) A burro was exposed to a mock-up of the liquid assembly to determine the relationship of Na^{24} activity in the blood and neutron dose. The ratio of γ-dose, D_γ, to neutron dose, D_n, for this particular type of critical assembly was also determined with the mock-up experiment (see Section IV). (3) A parallel program calculated these same two quantities. The calculations, described in Sections II and III, proved to be extremely helpful since they provided a basis for checking the experimental procedure at various stages.

II. SODIUM ACTIVATION OF HUMAN BLOOD

The (n,γ) process in Na^{23} gives rise to Na^{24} which has a half-life of 14.8 hr and emits a 1.38 MeV γ-ray in cascade with a 2.76 MeV

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

† The personnel security badge did contain strips of indium which proved invaluable in separating the exposed from the non-exposed persons.

‡ Because of complex physiological factors involved in the interpretation of P^{32} activity in urine, this work will deal almost entirely with the interpretation of Na^{24} activity in blood. Both P^{32} and Na^{24} in urine were analyzed by L. B. Farabee, ORNL. In the case of Na^{24} in urine, it appears possible to correlate activity with neutron dose, particularly if the first void after exposure is not used and if samples are collected before new sodium is given to the exposed individual. In the case of P^{32}, the difficulties reported in Ref. 3 were encountered.

γ-ray per Na^{24} decay. One expects that the cross-section for this process will follow a $1/v$ behavior from the thermal region to about 300 eV. The capture contribution of the resonance at 2.94 keV has been estimated by GOLDSTEIN[4]. He finds a very narrow resonance with a maximum cross-section at resonance of only 1 barn. It is easy to show that the number of captures occurring in this resonance is negligible compared with those occurring at thermal energies for slowing-down spectra typical of hydrogenous media. GOLDSTEIN notes that some uncertainty exists in his estimate of the resonance cross-section and that further experimental work will be necessary to resolve the uncertainty. However, it is expected that the neglect of epithermal activation of Na^{23} is not serious and this approximation will be made throughout this paper.

The human body is several mean free paths thick for fast neutrons. Consequently, the probability that a fast neutron will be captured as a thermal neutron is not very sensitive to its initial energy. Thus the body is roughly equivalent to the "long counter"[5] used so much in neutron flux measurements. The activation of elements in the blood should be a valid measure of the average thermal flux throughout the body and hence of the total flux incident at all energies.

This assumption is basic to the procedure described below for calculating the dose to those exposed in the nuclear excursion. The Na^{24} activation ($\mu c/cm^3$) of blood in the human body will be calculated under the assumption that the body is equivalent to a cylindrical phantom of 15 cm radius composed of soft tissue.

Calculations have been made by SNYDER and NEUFELD[6] of the depth distribution of thermal neutrons in an infinite slab of tissue-equivalent material 30 cm thick due to a monoenergetic broad beam irradiation by normally incident fast neutrons. The thermal neutron flux distribution is proportional to the curves labelled T_p in Figs. 1 through 11 of SNYDER and NEUFELD[6]. From these curves one may obtain the fraction of those neutrons entering the phantom which are captured at thermal energy. This fraction is plotted in Fig. 1 as a function of the incident neutron energy. Also plotted are values for the fraction $\xi(E)$ captured in a right cylinder of 15 cm radius, obtained from the slab data by making approximate corrections for thermal and fast leakage through the surface of the cylinder. The fast leakage was calculated by simply averaging over the path length distribution in a cylinder irradiated normal to its axis, taking the attenuation kernels for plane slab penetration from the data given by SNYDER and NEUFELD[6]. The thermal leakage was determined by applying one velocity diffusion theory to those neutrons which have entered the thermal region while in the cylinder. More accurate calculations of the capture fraction for a cylindrical phantom are

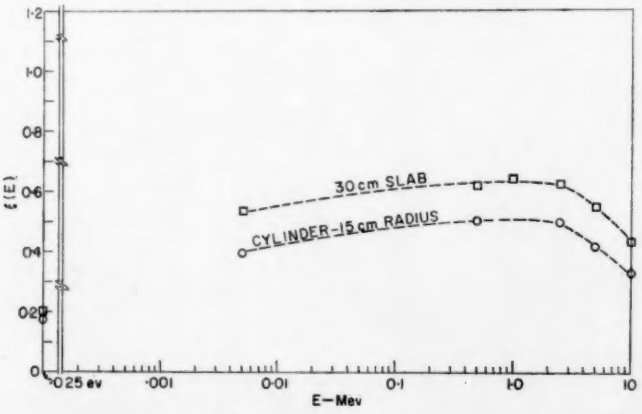

FIG. 1. Thermal neutron capture probability in a phantom.

being carried out by W. S. Snyder, using stochastic methods.

One may obtain an estimate of the Na^{24} activity, engendered in a cylindrical phantom by a beam of fast neutrons of energy E incident normal to the axis of the cylinder, by noting that the number of neutrons in the energy interval dE at E striking the cylinder is $A_p t\phi(E)dE$, where $\phi(E)dE$ is the fast neutron flux (n/cm^2-sec) in the energy interval dE at E, t is the irradiation time, and A_p is the area of the cylinder projected on a surface normal to the beam. Of the total number striking, a fraction $\xi(E)$ will be captured in the phantom and the fraction of these which will undergo capture in the $Na^{23}(n,\gamma)Na^{24}$ process is Σ_{Na}/Σ_t, where Σ_{Na} is the macroscopic absorption cross-section of Na^{23} in tissue, and Σ_t is the total absorption cross-section. Hence the total number of Na^{24} atoms appearing in the phantom as a result of irradiation will be given by

$$A_p \frac{\Sigma_{Na}}{\Sigma_t} t \int_0^\infty \xi(E)\phi(E)\, dE.$$

If the volume of the cylinder is V and the disintegration constant of Na^{24} is λ(μc/atom of Na^{24}), then the total Na^{24} activity per unit volume, $\mathscr{A}$, induced in the phantom by the flux $\phi(E)$ will be

$$\mathscr{A} = (A_p/V)\,(\Sigma_{Na}/\Sigma_t)\,\lambda t \int_0^\infty \xi(E)\,\phi(E)\,dE. \quad (1)$$

The total fast neutron first collision dose, D_n, associated with the beam is given by $D_n = \int_0^\infty D_0(E)t\phi(E)\,dE$, where $D_0(E)$ is the first collision soft tissue dose in rads per unit incident flux.[7]

Hence $(D_n/\mathscr{A})$, the total first collision fast neutron dose received by the phantom per unit activation of Na^{23} due to a flux distributed in energy like $\phi(E)$, is given by

$$\frac{D_n}{\mathscr{A}} = \frac{\int_0^\infty D_0(E)\phi(E)\,dE}{\frac{A_p}{V}\frac{\Sigma_{Na}}{\Sigma_t}\lambda\int_0^\infty \phi(E)\xi(E)\,dE}. \quad (2)$$

The V/A_p applies to a cylinder with a 15 cm radius. The cross-section Σ_t was calculated from the elemental abundance in the standard man[7] and is equal to 0.02339 cm^{-1}, assuming unit density for the body. Σ_{Na} was calculated using an average[8] value of 83 mequiv/l. or 1.906 mg of Na^{23} per cm^3 of whole blood and an absorption cross-section of 0.536 barns.[9] One finds, approximately

$$\frac{D_n}{\mathscr{A}} \cong 5.9 \times 10^{13} \frac{\int_0^\infty D_0(E)\phi(E)\,dE}{\int_0^\infty \xi(E)\phi(E)\,dE}$$

(rad/μc per cm^3). (3)

It has been assumed throughout all of the work reported here that the spectrum of neutrons and γ-rays in the neighborhood of the reactor is independent of distance from the critical assembly. This is thought to be a reasonable assumption for the range of distances involved. Then one may write that the flux $\phi(E)$ at a distance R from the assembly is given by $\phi(E,R) = N(E)/4\pi R^2$, where $N(E)$ is the number of neutrons with energy E per unit energy interval leaking from the assembly.

Clearly, the replacement of the body by a cylindrical phantom for the purpose of calculation is accompanied by some uncertainties. One must assume (a) that the blood is uniformly distributed throughout the body and hence takes on a sensible average activation and (b) that the geometrical differences between the human body and the cylindrical phantom are negligible with respect to the thermal capture of incident fast neutrons. It is believed that the approximations involved will not lead to serious error in the result.

The Na^{24} method of determining the fast neutron dose is most useful in cases where the number of thermal neutrons is not large compared with the number of fast neutrons. If the number of thermals is comparatively large, the bulk of the activation will be due to neutrons which do relatively little damage to the body and the accuracy of the method will suffer. However, if the spectral distribution of neutrons is well known, one may still derive an accurate dose value. In most cases of interest to the health physicist the thermal flux component will be small enough that the Na^{24} activation may

be related to the fast neutron dose with good accuracy.

III. CALCULATION OF D_γ/D_n FOR LIQUID ASSEMBLIES

Following the accident it seemed desirable to estimate the leakage spectra of the neutrons and the γ-rays. The overall accuracy of the methods employed in the case of the fast neutron leakage is not easy to assess in view of the rather complicated geometry of the assembly and surrounding materials but the calculated spectrum seems to agree reasonably well with the measured spectrum. A more accurate calculation is planned, in which stochastic methods will be employed.

The neutron and gamma ray leakage spectra were calculated under the assumption that the assembly could be considered spherical and the distribution of nascent fission neutrons in the assembly could be adequately represented by the fundamental spatial mode for a spherical reactor, i.e. proportional to $1/r \sin \pi r/r_0$, where r is the distance from the center of the assembly and r_0 is the radius of the equivalent sphere at the prompt critical stage (24.25 cm). The number of neutrons and γ-rays escaping the assembly was calculated by using infinite medium penetration data for point isotropic sources and by integrating these data over the volume source distribution assumed. This approximate procedure of employing infinite medium attenuation data is dictated by the lack of information relative to the bounded medium. The error involved in this procedure is believed to be small.

The details of calculation of the neutron escape spectrum will now be described. The escape of high energy neutrons ($E > 0.5$ MeV) from the assembly was carried out using infinite medium penetration data for fission neutrons in water.[10] One fits $\psi(E,r)$, the flux of neutrons of energy E at a distance r in water from a unit point source of fission neutrons, by an expression of the form,

$$\psi(E, r) = \frac{1}{4\pi r^2} \sum_i A_i(E) \exp\left(-r\mu_i(E)\right) \quad (4)$$

where the constants $A_i(E)$ and $\mu_i(E)$ are determined from an empirical fit to the data.

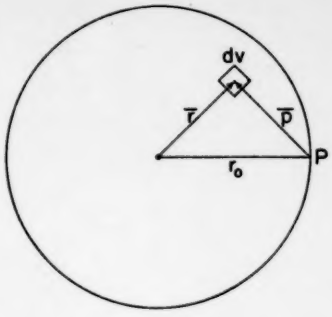

FIG. 2. Geometry used in calculating sphere transmission factors.

In all cases considered, only a few terms in the series were needed to obtain a good fit to the data. Then, one needs to calculate the leakage fraction through the surface at r_0 for a general attenuation kernel proportional to $e^{-\mu r}/4\pi r^2$ in order to determine the leakage fraction for the normalized distribution $\mathscr{S}(r)$ inside the sphere. The leakage fraction $\mathscr{L}$ through the surface is given by

$$\mathscr{L} = 4\pi r_0^2 \int dv \frac{e^{-\mu \mathbf{p}}}{4\pi \mathbf{p}^2} \cos\varphi \mathscr{S}(r) \quad (5)$$

where $\mathbf{p}$ is a vector to the volume element dv from the point P on the surface and $\cos\varphi$ is the angle between $\mathbf{p}$ and the inward normal to the surface at P (see Fig. 2). The integration is carried out over the interior of the sphere. Integrating over the volume by first varying $\mathbf{p}$ while holding r constant and then finally integrating over r, one finds

$$\mathscr{L}(\mu r_0) = \frac{1}{4r_0^2} \int_0^{r_0} r\, dr \mathscr{S}(r) \Big\{ [r_0 + r] \times E_2[\mu(r_0 - r)] - [r_0 - r] \times E_2[\mu(r_0 + r)] + \frac{1}{\mu r_0} \times [\exp(-\mu(r_0 - r)) - \exp(-\mu(r_0 + r))] \Big\} \quad (6)$$

where

$$E_2(x) = \int_1^\infty \frac{du}{u^2} e^{-xu}.$$

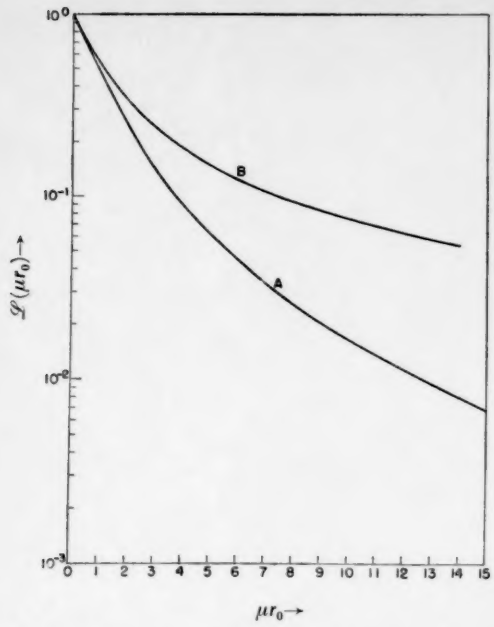

FIG. 3. Sphere transmission factors for an exponential attenuation kernel. Curve A is for a source distributed as $1/4\pi r_0^2 r\ (\sin \pi r/r_0)$. Curve B is for a uniform source distribution.

The leakage fraction $\mathscr{L}_A(\mu r_0)$ for the normalized critical mode

$$\mathscr{S}(r) = \frac{1}{4\pi r_0^2 r} \sin \frac{\pi r}{r_0}$$

was evaluated by numerical integration and is given as curve A of Fig. 3.

Then the leakage spectrum $N(E)$ for neutrons of energy E is given by

$$N(E) = \sum_i A_i(E) \mathscr{L}_A[r_0 \mu_i(E)]. \qquad (7)$$

The attenuation data were fitted at several energies and the values of A_i and μ_i were substituted in this equation. The result is shown in Fig. 4. Note that the normalization in this figure refers to one fission in the core rather than to one neutron so that the data obtained from this formula have been multiplied by 2.47 neutrons/fission to obtain $N(E)$. Since the results of ARONSON *et al.*[10] are valid only for relatively high energies, it was necessary to

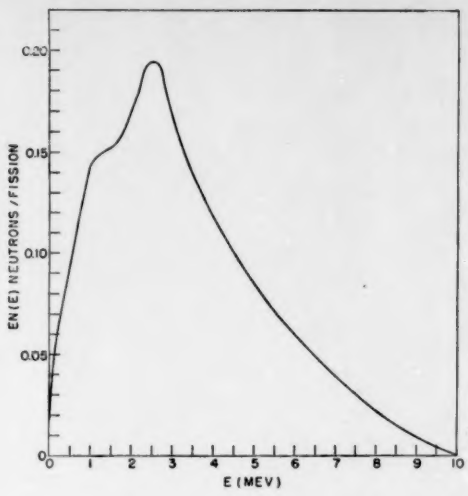

FIG. 4. Neutron leakage spectrum.

supplement them by independent calculations in the low energy region.

A multigroup, multiregion reactor analysis IBM-704 code[11] applicable to water moderated systems was available and was used for this purpose.* Since this code is limited to a constant extrapolation distance (independent of energy group), two independent calculations were made, one with an extrapolation length found from an overall group average, and the other with a value appropriate to the high energy region. The leakage in the high energy region predicted by the second calculation was not inconsistent with the results obtained from the infinite medium attenuation kernels. However, since the leakage spectrum was given rather incompletely in this range of energies by this particular multigroup code and since the diffusion approximations are rather suspect at these energies, the attenuation kernel results were employed. The results of the first multigroup calculation of the leakage at low energies were used. The resulting estimate of the composite spectrum is shown in Fig. 4. The ordinate in this figure is $EN(E)$, where $N(E)dE$ is the total number of neutrons in dE at E leaking

* The writers are indebted to Mr. V. E. ANDERSON of the Numerical Analysis Group of the Oak Ridge Gaseous Diffusion Plant for carrying out this portion of the calculation.

Table 1. *Relative flux density of neutrons for various energy regions*

Energy range	$(\text{Neutrons/cm}^2)_{\text{exp.}}$	Normalization	
		$(\text{Neutrons/cm}^2)_{\text{exp.}}$	$(\text{Neutrons/cm}^2)_{\text{theor.}}$
Thermal	0.90×10^{11}	0.45	0.08
Total fast	2.0×10^{11}	1.00	1.00
0.5 keV to 0.75 MeV	0.25×10^{11}	0.125	0.30
0.75 MeV to 1.5 MeV	0.70×10^{11}	0.35	0.204
1.5 MeV to 2.5 MeV	0.58×10^{11}	0.29	0.246
>2.5 MeV	0.50×10^{11}	0.25	0.250

from the assembly per fission occurring in the interior. The total dose, $\int_0^\infty \mathscr{D}_0(E)N(E)\,dE$, in this spectrum is found to be 1.08×10^{-9} rad-cm^2/fission, and the ratio $\mathscr{D}_n/\mathscr{A}$ is 1.95×10^5 rad-cm$^3/\mu$c [equation (3)].

As stated above, it is assumed throughout this paper that the spectrum of escaping neutrons does not depend appreciably upon distance from the assembly, so that in equation (3) for $D_n/\mathscr{A}$ one may substitute $N(E)$ for $\phi(E)$. Although this can be only approximately true, since there is clearly an appreciable amount of scattering and degradation of the spectrum in the materials of the room, one expects that this will not lead to serious error in the dose estimates. Note that the thermal component of the calculated neutron spectrum is smaller than one finds experimentally (Table 1). The higher experimental value is probably explainable in terms of room scattering and is not inconsistent with the results of HURST, MILLS and REINHARDT* on the leakage spectrum of neutrons from the Godiva reactor.

Three sources of γ-radiation were considered in the analysis: (a) prompt γ-radiation emitted directly from the fission process, (b) capture γ-radiation resulting from (n,γ) reactions taking place within the assembly, and (c) delayed γ-radiation from the fission products contained within the assembly. The spectral distribution of the prompt γ-rays, $\Gamma(E)$, was obtained directly from the recent work of MAIENSCHEIN *et al.*[12] and is shown in Fig. 5. The spatial distribution of this source is determined by the spatial distribution of the fission events and is assumed to be proportional to $1/r \sin(\pi r/r_0)$. The capture γ-sources were obtained from the following equation:

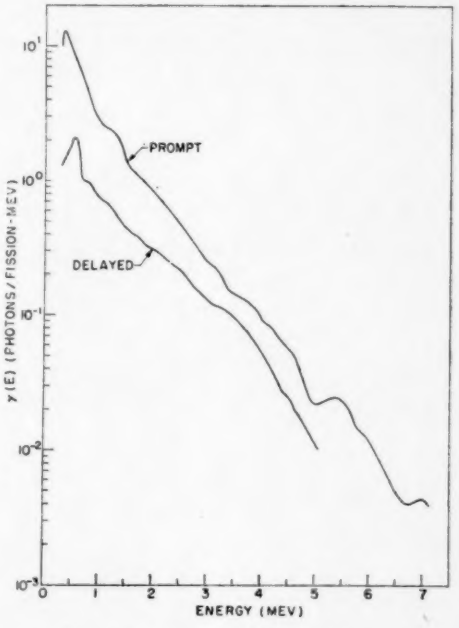

FIG. 5. Prompt and delayed γ-spectra.

$$\Gamma_j = \nu(1 - f_c)\left(\frac{\sigma_{rc}}{\sigma_a}\right)_j \left(\frac{E_c}{E_\gamma}\right)_j \frac{(\Sigma_a)_j}{\Sigma_t} \qquad (8)$$

* G. S. HURST, W. A. MILLS and P. W. REINHARDT, unpublished data on neutron leakage spectrum from the Godiva reactor at LASL.

Table 2. Capture γ-ray spectra

Element	E_c (MeV/capture)	E_γ (MeV/photon)	S (photons/fission)
Hydrogen	2.2	2.2	0.478
Uranium	6.8	3.0	0.427
		1.0	0.171

where

Γ_j = photons of energy E_γ released from the jth element per fission;

ν = neutrons born per fission (2.47);

f_e = neutron escape fraction (0.271);

σ_{rc} = microscopic radiative capture cross-section;

σ_a = microscopic absorption cross-section;

E_c = energy release per neutron capture in form of γ-rays of energy E_γ;

Σ_a = macroscopic absorption cross-section;

Σ_t = total macroscopic absorption cross-section of the assembly.

Uranium and hydrogen were the only elements in the assembly which contributed appreciably to the capture γ-dose. For these elements the values for E_c and E_γ were obtained from GLASSTONE[13]. In the case of uranium these values are not well known but the total neutron binding energy of 6.8 MeV is accounted for correctly by the proposed γ-spectrum. The calculated values for the capture γ-sources are shown in Table 2. As in the case of the prompt radiation, the capture γ-rays are released essentially simultaneously with fission so that the spatial distribution again follows that of the fission events.

It was not necessary to determine the complete leakage spectrum of prompt γ-rays from fission and γ-rays from capture. One is interested primarily in the γ-dose escaping the system. The calculation of the leakage dose for prompt and capture γ-rays was carried out by first computing the variation of dose $\mathbb{D}(r)$ with distance in an infinite medium having the composition of the assembly due to a unit point isotropic source having the indicated distribution of energy. Thus,

$$[\mathbb{D}(r)]_{\text{prompt}} = K\int_0^\infty E\Gamma(E)\mu_a{}^t(E)B_d(E,\mu r) \times \frac{\exp(-\mu(E)r)}{4\pi r^2}\,dE \qquad (9)$$

where

$\mathbb{D}(r)$ = γ-dose at r cm from a unit source (rad/fission);

$K = 1.6 \times 10^{-8}$ rad/MeV-gm^{-1};

$\Gamma(E)$ = number of γ-rays of energy E emitted per unit interval from one fission (photon/fission MeV);

$\mu_a{}^t(E)$ = energy absorption coefficient for tissue dose (cm^2/g);

$B_d(E, \mu r)$ = dose buildup factor;[14]

$\mu(E)$ = attenuation coefficient of water for photons of energy E (cm^{-1}).

The integration was performed numerically using values of B_d obtained from the work of GOLDSTEIN and WILKINS[14]. $[\mathbb{D}(r)]_{\text{capture}}$ was evaluated in the same way except that a sum over the j capture photon groups was carried out. The computed curves for $[\mathbb{D}(r)]_{\text{prompt}}$ and $[\mathbb{D}(r)]_{\text{capture}}$ were then fitted by empirical formulas of the form used in the neutron calculation,

$$\mathbb{D}(r) = \frac{1}{4\pi r^2}\sum_i A_i \exp(-\mu_i r) \qquad (10)$$

and the dose, D_γ, escaping per fission was found from the equation

$$D_\gamma = \sum_i A_i \mathscr{L}_A(\mu_i r_0). \qquad (11)$$

The treatment of the delayed radiation presents somewhat more of a problem as both

the source strength and the energy distribution are strongly time dependent during the first few seconds following fission. The spectral distribution shown in Fig. 3, was obtained by normalizing the spectrum measured by MAIENSCHEIN *et al.*[12] at 6.2 sec to the integral over the first 15 sec of the measured decay spectrum. It was necessary to extrapolate to obtain values during the first second. The delayed γ-emitters were assumed to be homogeneously distributed throughout the assembly because of turbulence following the initial release of energy, hence it was necessary to obtain transmission factors for uniformly distributed sources of γ-rays. The leakage fraction, $\mathscr{L}_B(\mu r_0)$ for a normalized uniform distribution $\mathscr{S}(r) = \dfrac{3}{4\pi {r_0}^3}$ may be written explicitly as

$$\mathscr{L}_B(\mu r_0) = \tfrac{3}{8}\,(\mu r_0)^{-3} \times [2(\mu r_0)^2 - 1 + (1 + 2\mu r_0)\exp(-2\mu r_0)] \quad (12)$$

which may be shown by carrying out the integration indicated in equation (6). This result is shown as curve B of Fig. 3.

The movements of the exposed persons subsequent to the initial excursion influenced the amount of γ-radiation which they received. In the dose analysis it was assumed that each person exposed was exposed to a single nuclear excursion but that his individual actions in the few seconds following the excursion resulted in different γ-neutron dose ratios. This results in a delayed γ-dose slightly larger than that obtained by assuming a constant power assembly delivering the same total fission energy. The following three cases treated were based upon the actions of each of the individuals as determined from personal interviews.[15]

Case I (*employee* A). This employee was exposed to the full complement of prompt and capture γ-radiation but, since it is believed that he left the area first, it was assumed that only the first 5 sec of the delayed γ-spectrum contributed to his dose.

Case II (*employee* E). This employee was also exposed to the prompt and capture γ-radiation source, but the γ–neutron ratio was adjusted to take into account an assumed 15 sec exposure to the delayed γ-spectrum and, further, that his exit from the area took him to within 10 ft of the assembly. This case gives the largest γ–neutron dose ratio used in analysis and reflects the fact that this employee was apparently the last to leave the area. The assumed exit path resulted in a total γ-dose which was 8 per cent more than the dose of the constant distance 15 sec exposure case. This represents what is believed to be the maximum increase that could have occurred for any of the personnel.

Case III (*employees other than* A *and* E). These employees were assumed to have received the full contribution of prompt and capture γ-radiation, but, like case I, the delayed γ-rays were received at a constant distance and, like case II, the first 15 sec of the delay spectrum was effective.

The escaping dose from delayed γ-rays which was received in the three cases considered was determined by the same method used for the prompt and capture γ-dose calculation. The dose $[\mathbb{D}(r)]^{(J)}_{\text{delayed}}$ was found from equation (9) in which the portion of the delayed spectrum $[\Gamma(E)]^{(J)}_{\text{delayed}}$ seen by the Jth case was employed. $[\mathbb{D}(r)]^{(J)}_{\text{delayed}}$ was then fitted by an empirical expression having the form of equation (10) and the escaping dose $[D_\gamma]^{(J)}_{\text{delayed}}$ seen by case J was calculated from the equation:

$$[D_\gamma]^{(J)}_{\text{delayed}} = [\sum_i A_i \mathscr{L}_B(\mu_i r_0)]^{(J)}.$$

The results for the dose multiplied by $4\pi r^2$ are shown in Table 3.

IV. DOSE MEASUREMENTS WITH A MOCK-UP OF THE CRITICAL ASSEMBLY

To establish experimentally the relationship between the total (neutron + γ) first collision dose and the blood Na activation, a mock-up of the critical assembly was constructed and operated* as a low power reactor in two experiments. Fig. 6 shows the actual reactor tank surrounded with a large tank which rested on a concrete floor. Table 4 gives details of the mock-up assembly as compared to the actual critical assembly. The first experiment was designed to measure the ratio of γ-dose D_γ, to neutron dose D_n, i.e. D_γ/D_n. The assembly was operated at a power of about 6 W for 11 min. The γ-dose rate at 6 ft from the center of the assembly, point A in Fig. 6, was measured with an ionization

* This phase of the work was carried out at the ORNL criticality facility.

Table 3. Results of γ-dose calculation

Case no.	Prompt (rad-cm²/fission)	Capture (rad-cm²/fission)	Delayed (rad-cm²/fission)	Total (rad-cm²/fission)	D_γ/D_n
I	2.06×10^{-9}	6.4×10^{10}	4.0×10^{-10}	3.10×10^{-9}	2.87
II	2.06×10^{-9}	6.4×10^{-10}	9.4×10^{-10}	3.64×10^{-9}	3.37
III	2.06×10^{-9}	6.4×10^{-10}	6.4×10^{-10}	3.34×10^{-9}	3.09

Table 4. Comparison of the mock-up critical assembly with the actual critical assembly

	Actual critical assembly	Mock-up critical assembly
Diameter (in.)	22	20
Height (in.)	9.2	15
U^{235} conc. (g/l.)	37.4	25.9
Total U^{235} (kg)	2.1	2.00

chamber having carbon walls and CO_2 gas.* This ionization chamber was calibrated with Co^{60} γ-radiation to read the first collision tissue dose in rads. Measurements of the neutron dose rate were carried out at the above mentioned position with the Radsan fast neutron dosimeter,[16] which uses the absolute proportional counter.[17] These two sets of measurements gave a value of 3.3 for the ratio D_γ/D_n.

The ratio $D_\gamma/D_n = 3.3$ applies for the reactor which has been running at constant power for approximately 3 min, the time after start-up at which the measurement was made. In order to compare this result with those given in Section III, this ratio must be corrected to correspond to the case where (a) the critical assembly is assumed to give an instantaneous pulse of radiation, and (b) only the first 15 sec of the decay γ-radiation is assumed to be effective. When the experimental results of Maienschein *et al.*[12] for the spectrum of the decay γ-rays are used, the experimentally determined ratio, D_γ/D_n, becomes 2.8 compared to the calculated ratio of 3.09 given in Table 3.

In the second experiment with the mock-up critical assembly, a burro was exposed in such a way that a point on his plane of symmetry coincided with the point *A* where the neutron dose was measured in the first experiment. The burro was chosen as the experimental animal because (a) he is a large

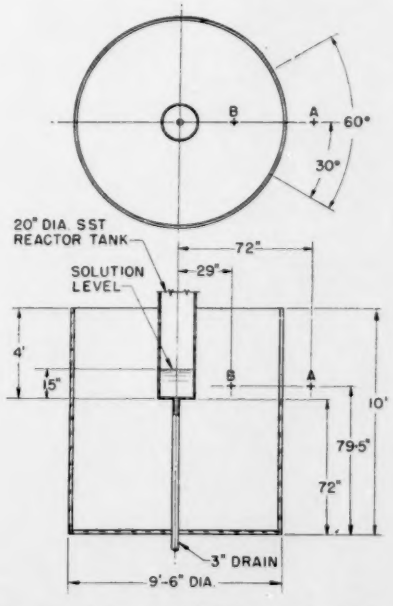

FIG. 6. Schematic of mock-up experiment.

* This type of ionization chamber has been used extensively by the Neutron Physics Division, ORNL, in their shielding program. For a description, see L. H. Ballweg and J. L. Meem, *Standard Gamma-Ray Ionization Chambers for Shielding Measurements*. ORNL-1028.

Table 5. Blood sodium for individuals exposed and for experimental burro

Exposed individuals	Blood sodium (mg/ml serum)
A	3.2
B	3.2
C	3.2
D	3.1
E	3.2
Experimental burro	3.1

animal comparable in size to man, and (b) the amount of sodium per gram of blood serum is approximately the same for burro and man. Table 5 shows the concentration of sodium in blood serum for the burro and for some of the exposed individuals.*

The second experiment was operated at a power of about 300 W for 42 min which gave the burro a fast neutron first collision dose of 48 rads.† Blood samples were drawn from the burro and counted for Na^{24} as described in the next section. From these data the value of $\mathscr{A}/D_n$ for this particular neutron spectrum was determined to be 6.0×10^{-6} μc of Na^{24} per ml of blood and per rad of neutron dose, compared to 5.14×10^{-6} μc obtained by using the methods given in Sections II and III. Note that the results calculated in Section II apply to the case where a model of a man is the irradiated object. The close agreement of these two values is further indication that the burro is a good phantom for a man.

Also, in the second experiment a series of threshold detectors[18,19] was used to obtain information on the fast neutron spectrum. The neutron detectors [Au (Au + Cd), S^{32} and Pu^{239}, Np ^{237} and U^{238} in 2.2 g/cm^2 B^{10}] were exposed at approximately 29 in. from the center of the liquid assembly, point *B* in Fig. 6. The total flux of neutrons received during the 42 min period for various energy ranges is shown in Table 1. The table also shows a comparison with the calculations given in Section III. Using the flux values measured at point *B* the first collision neutron dose can be calculated[18] at the burro exposure position (point *A*). A sulfur disc was fixed on the side of the burro facing the assembly. The measured value of the flux was corrected for distance and for flux build-up‡ due to the burro to obtain the flux which would have existed at point *A* had the burro not been there. The first collision neutron dose at point *A* was then determined from the ratio of the neutron flux at point *A* to that at point *B* and the neutron dose calculated for point *B*. Thus the threshold detector data when suitably corrected to refer to point *A* gives a first collision neutron dose at point *A* equal to 42 rads compared to 48 rads using the proportional counter method.

The value of the thermal neutron flux in Table 1 shows that the dose due to thermal neutrons is negligible compared to the fast neutron dose. However, the magnitude of the thermal neutron flux is of interest because of another factor. In Fig. 1 it is seen that the probability that a thermal neutron entering the human body will produce Na^{24} is about 0.4 as large as the probability that a fast neutron will become thermalized and produce a Na capture in the body. Thus it would be expected that for the spectrum shown in Table 1 only about 15 per cent of the Na^{24} activity would be produced by thermal neutrons. This fact is of considerable practical importance since it implies that it is not necessary to consider the spatial distribution of thermal neutrons in detail.

V. ASSIGNMENT OF DOSE VALUES TO THE EXPOSED INDIVIDUALS

The blood samples collected from the individuals were counted with a 2 × 4 in. NaI crystal (Fig. 7). Polyethylene bottles (4 fluid oz capacity) containing the whole blood were placed on the top of the crystal. The horizontal lines indicate liquid levels for 50 and 100 ml. Counting was done with a discriminator set to accept γ-rays above 0.66 MeV and 2.0 MeV. The equipment was calibrated with 50 and 100 ml solutions of Na^{24} in water. The counting geometry for Na^{24} as well as the counter background (using a 4 in. lead shield) is shown in Table 6 for the two bias values and for 50 and 100 ml samples.

A few hours after exposure, 100 ml blood samples were taken from some of the individuals, and this was counted as whole blood without the use of an anticoagulant. A second set of blood samples was

* These measurements were made by Dr. C. H. Steffee, Oak Ridge Hospital, using the flame photometer technique. Although it was not possible to obtain whole blood sodium values at the time the experimental work was done, a subsequent evaluation of whole blood sodium for unirradiated burros showed results not inconsistent with values to be expected in normal man.

† The amount of exposure was determined with a U^{235} fission chamber which had been calibrated in terms of fast neutron dose in the first experiment.

‡ The depth dose curves taken from W. S. Snyder, *Brit. J. Radiol.* **28,** 342 (1955), and *NBS Handbook* 63, November 1957, were used as a basis of correction.

Table 6. Na^{24} *counting geometry and background for* 2 *by* 4 in. NaI *crystal at two bias levels*

Bias (MeV)	Geometry (50 ml) (%)	Geometry (100 ml) (%)	Background (counts/min)
0.66	17.7	13.3	250
2.0	5.9	4.3	115

Table 7. Na^{24} *activation and dose values for exposed personnel*

Exposed individuals	Na^{24} (μc/ml)	Experimental neutron dose (rad)	Theoretical neutron dose (rad)	Experimental* γ-dose (rad)	Theoretical† γ-dose (rad)	Total experimental dose (rad)
A	5.8×10^{-4}	96	113	269	349	365
B	4.3×10^{-4}	71	83	199	256	270
C	5.4×10^{-4}	89	104	250	321	339
D	5.2×10^{-4}	86	100	241	308	327
E	3.7×10^{-4}	62	72	174	223	236
F	1.1×10^{-4}	18	21	50.5	64.9	68.5
G	1.1×10^{-4}	18	21	50.5	64.9	68.5
H	0.36×10^{-4}	6.0	7.0	16.8	21.6	22.8
Burro	2.9×10^{-4}	48				

* Using $D_\gamma/D_n = 2.8$.
† Using $D_\gamma/D_n = 3.09$.

taken about 20 hr later, but this time only 50 ml was used and heparin was added to prevent clotting. This set of data was more nearly uniform than the first and it is the basis of the final dose values reported. The blood samples were counted for several days to establish that Na^{24} was the primary isotope being counted. Very early counting showed some evidence of K^{42} in the case when the counting bias was 0.66 MeV.

Table 7 shows the final results for the Na^{24} activity (extrapolated to the time of exposure) for the exposed personnel as well as that for the experimental burro. Blood samples (100 ml) from the burro, receiving a neutron dose of 48 rads, were counted‡ in the same way as described above for the exposed individuals.

Table 7 shows the first collision neutron dose, γ-dose, and total dose obtained from the experimental data by direct comparison of Na^{24} activities in the human blood with that of the burro blood, using the known fast neutron dose given to the burro and the experimentally measured ratio, D_γ/D_n. Also shown in Table 7 are the dose values obtained by calculations, Sections II and III.§

The counting methods described above are very sensitive. For example, it is seen by using data from Tables 6 and 7 that 1 rad of neutrons give a count rate due to Na^{24} equal to 40 counts/min (at zero time) for 50 ml burro blood when the counting bias is 2.0 MeV. Thus, since the counter background rate is 115 counts/min, a dose of 1 rad can be detected. With minor improvements in counting techniques, it should be possible to determine doses as low as 100 mrads with the Na^{24} method.

‡ For the first few hours Cl^{38} competed with Na^{24}, especially at the 0.66 MeV bias.

§ The calculated values given in the table differ from the corresponding values in report Y-1234, *Accidental Excursion at the Y-12 Plant*, (4, August 1958). The inadvertent use of a high value of the concentration of Na^{23} in whole blood gave calculated values which were too low.

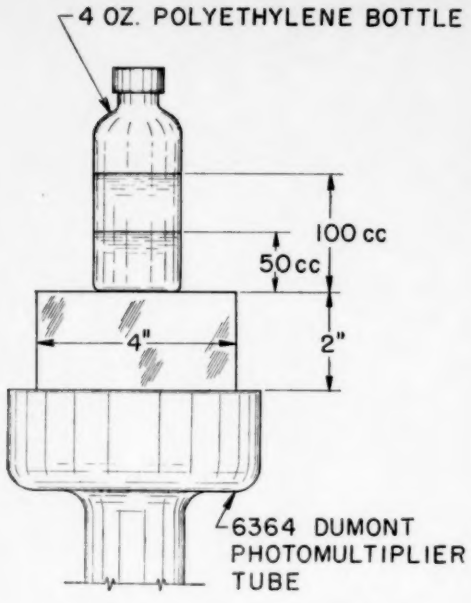

FIG. 7. Geometry of the blood counter.

VI. DISCUSSION OF ERRORS

As mentioned in Section I, comparison of calculations with experiment leads to independent tests of some of the assumptions made in the experimental program. The various sources of error in the experimental approach and the extent to which comparison of theory and experiment helps to decrease the uncertainty involved in individual assumptions will now be summarized. The assumptions made in the experimental program are:

(1) The actual radiation spectrum is duplicated with the mock-up reactor.

(2) Na^{24} activity in blood of the exposed person is approximately proportional to the first collision neutron dose, i.e. the Na^{24} activity resulting from capture of thermal neutrons coming from the reactor constitutes a correction factor.

(3) The relationship of Na^{24} activity in the blood of a burro, exposed to the radiation from the mock-up reactor, to the first collision dose is valid for a man exposed to the actual radiations.

(4) The γ-dose for each of the exposed persons may be established by determining the ratio D_γ/D_n for the mock-up facility.

Assumption (1) was checked by measuring the neutron leakage spectrum from the mock-up with threshold detectors. The neutron leakage spectrum from the actual assembly was calculated. Table 1 shows comparison of these two spectra. The γ-spectrum for the actual assembly was calculated with what is thought to be reasonable accuracy. Errors in dose due to the difference in spectra were thought to be negligible.

Assumption (2) may lead to error for some of the cases. It was shown by calculation (see Fig. 1) that thermal neutrons are less effective than fast neutrons in producing Na^{24} in the blood. Measurements of the thermal neutron flux for the exposed burro combined with this result show that only about 20 per cent of the Na^{24} is due to thermal neutrons coming directly from the reactor. However, for those persons at large distances from the reactor, the Na^{24} method may lead to overestimates of the dose. This point deserves further exploration.

Assumption (3) was checked by comparison with calculation. The relationship of neutron dose to Na^{24} activity was calculated for a man exposed to the neutron spectrum, found from calculations on the actual assembly, and compared to the value for a burro exposed to the mock-up. The value of rad per μc per ml of blood was 20 per cent higher for the calculated case.

Assumption (4) has already been considered in detail in Section III. Very little error results from this assumption.

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ACCIDENTAL RADIATION EXCURSION AT THE OAK RIDGE Y-12 PLANT*—IV

PRELIMINARY REPORT ON CLINICAL AND LABORATORY EFFECTS IN THE IRRADIATED EMPLOYEES

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(*Received* 7 *April* 1959)

Abstract—The patients exposed to irradiation in the Y-12 accident have been studied and followed at the ORINS Medical Division Hospital. The five patients with the higher dose levels were kept in the hospital for several weeks following exposure. Epilation began on the seventeenth day. Minimal clinical signs of a hemorrhagic tendency were seen around the twenty-fifth to thirtieth days. Antibiotics were not used prophylactically. A few minor infectious episodes occurred, but were easily managed. Hematologic studies showed the characteristic pattern of changes expected after total-body irradiation. Spontaneous recovery of blood values occurred and the patients have remained in good general condition following the accident.

To clinicians the accident that occurred at Y-12 on 16 June 1958, was of unusual interest and concern. The injury was one of whole-body penetrating irradiation uncomplicated by superficially deposited or internally ingested radioisotopes and uncomplicated by any pre-existing illness in the persons exposed. The dose range was sufficiently high to cause grave concern about the outlook of the exposed persons, and to raise the question of using a highly experimental form of treatment with bone-marrow transplantation.

CHRONOLOGIC OUTLINE

The patients studied included five men exposed to doses of about 300 rads and three with doses below 100 rads. The three men with low doses never showed more than slight or equivocal clinical and laboratory evidences of radiation effects. The present report is confined almost entirely to the five with higher doses. They were admitted to the hospital about 12 hr after the accident. All had nausea and four had vomiting. These symptoms began about 2 hr after exposure and persisted intermittently for about 48 hr. It is interesting that the one who did not vomit is believed to have been exposed to the second highest dose. The patients did not have fever after exposure. At this time there was no great concern over their immediate condition, which was good, but there was serious concern about the severity of the illness that might develop a few weeks after the exposure.

Biological estimates of radiation dosage were attempted during the first 3 days. In all five, blood lymphocytes fell in the first 48 hr to levels in the neighborhood of 1000/mm^3. Reticulocytes did not disappear from the blood films. Early bone-marrow studies showed no massive necrosis and only rather subtle changes in distribution of cell types and cell morphology. Three days after the accident, on the basis of an evaluation of all of the clinical, laboratory and dosimetric information available, a decision was made against immediate attempts at bone-marrow transplantation. At this point, relatives of the patients, who had been asked to come to Oak Ridge to serve as potential marrow donors, were released.

The patients were kept under careful observation. They appeared clinically well after the nausea had ceased. Beginning on the

* From the Medical Division, Oak Ridge Institute of Nuclear Studies, under contract with the United States Atomic Energy Commission.

seventeenth hospital day they were allowed daily home visits from 2 p.m. until 10 p.m. On the twenty-fourth day these "passes" were temporarily discontinued because the hematologic depression that had been expected was becoming quite prominent. Between the twenty-fifth and thirtieth days depression of white cell and platelet values was serious. At this time physical examination revealed definite evidence of a hemorrhagic tendency. Preparations were made for the immediate use of fresh whole blood or concentrated platelets, but spontaneous recovery occurred without these measures.

On the thirty-ninth hospital day the patients were again allowed to spend a part of each day away from the hospital, and on the forty-fourth day they were discharged. After discharge they became increasingly aware of weakness and especially of a feeling of tiredness or a dull ache, particularly of the thighs. These symptoms gradually decreased and about 6 weeks after discharge they returned to work. Since then they have been in good health without any major manifestations that could be attributed to the radiation exposure.

Fig. 1 helps to correlate the time relationships of hematologic and clinical developments.

ADDITIONAL CLINICAL FEATURES

Soon after the patients were admitted to the hospital, consideration was given to the advisability of using prophylactic antibiotics. It was decided that no antibiotics or anti-infectious chemical therapy would be given unless some definite evidence of infection developed. A few minor infections were encountered but most of them did not occur at a time when the granulocyte depression was pronounced. Patient *E* had a very small furuncle of the external auditory canal on the third day; it cleared up spontaneously. Patient *B* had a large furuncle, in the left gluteal region, which started on the tenth day and was finally incised and drained on the twenty-second day. There was no fever and no specific drug therapy was given for this lesion; however, the same patient developed pharyngitis and a right otitis media on the thirteenth day with a mild temperature elevation and was treated with tetracycline. Patient *C* developed a mild upper respiratory infection starting on the tenth day and was given no specific therapy. Patient *A* developed acute tonsilitis on the thirty-first day with a temperature of 38.6°C. He was treated with tetracycline.

All the patients had some evidence of hair loss starting about the seventeenth day, but in only two of them, patients *A* and *D*, was the hair loss so pronounced that it was obvious to any observer. These two patients developed almost complete baldness of a large rounded area on the back of the head. The other patients had some loss of hair from the head or body but no complete epilation. Three of the patients noticed some soreness of the scalp preceding and during the period of early hair loss.

Slight evidences of a bleeding tendency were apparent, and in one patient quite pronounced manifestations developed. These included petechiae, which were present in small numbers in patient *A* and in large numbers in patient *C*. Patients *A* and *E* had some bleeding of the gums on brushing, and three of the patients had a very small number of red cells in the urine. All these manifestations occurred between the twenty-fifth and twenty-eighth days when the thrombocytopenia was most pronounced.

All of the patients gained some weight (ranging from 3 to 13 lb) during their 44 days in the hospital. Three of them showed a slight early weight loss during the first few days of hospital care.

HEMATOLOGIC CHANGES

Fig. 1 shows the average blood values for the five high-dose patients during the first 60 days after the accident. Averaging of the values is considered justified because the radiation dose and the hematologic response were reasonably uniform in the whole group. Still, there were some differences, and the two patients, *A* and *C*, showed more profound changes in white cell, platelets and red cell values than did the other three. All the men had had previous blood counts done in the dispensary of the Y-12 plant. After the accident a blood count was done in that laboratory between 1½ and 2 hr after the exposure. Subsequent blood work was done in the laboratory of the

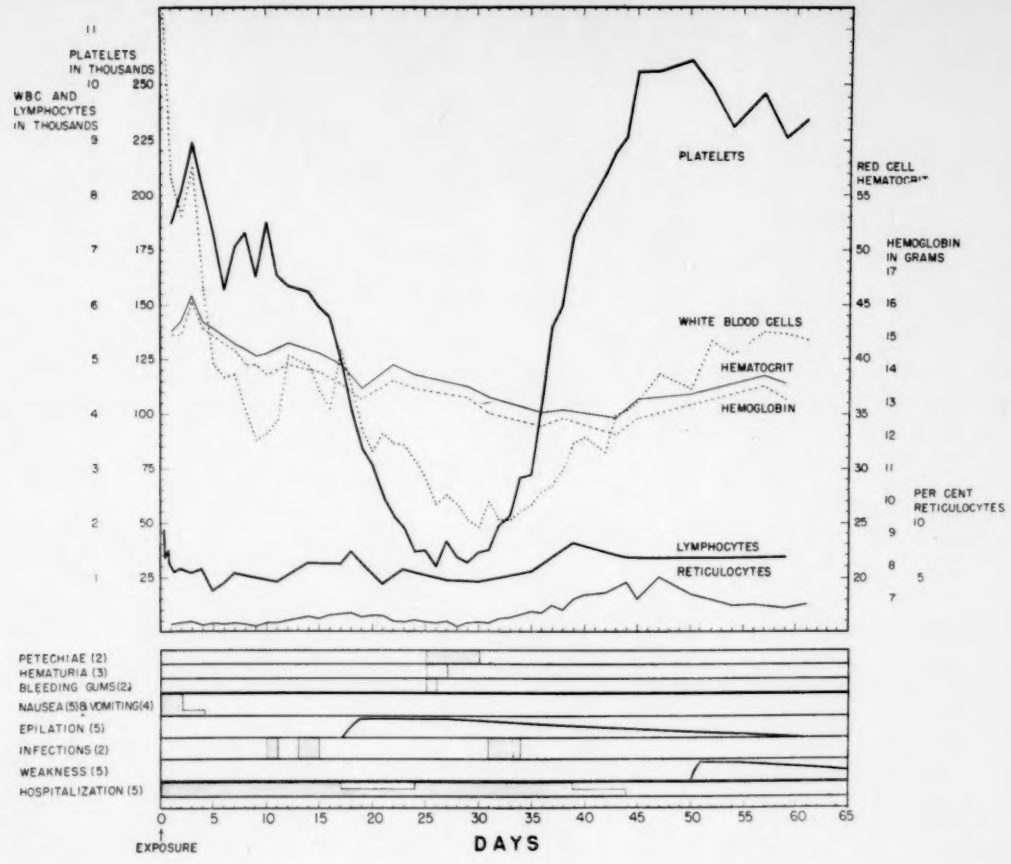

FIG. 1.

Medical Division. Blood films from the previous work at the Y-12 dispensary were reviewed so that criteria for differential counts could be uniform in the series. Fig. 1 shows only the work done after the accident. Base line counts, all within normal range, are not placed on the chart because they were all obtained many months earlier, but they are of importance in ruling out any pre-existing abnormality.

The most immediate change was a fall in lymphocytes. Two hours after the accident the average was around 1800, and by 48 hr it had fallen to slightly above 1000. Most of the fall in lymphocytes occurred within the first 48 hr, and after that the lymphocytes remained fairly uniformly depressed showing a slight rise after the thirtieth day. In two patients there was a rise in the total white cell count within 2 hr (unless for some unknown reason they had a leukocytosis at the time of exposure). This elevation in white count was chiefly accounted for by an increase in granulocytes. By the fourth day the leukocytosis had disappeared and the white counts were within low normal values. Between the tenth and eighteenth days there was apparently a slight transient rise in the total white count. Then there was a distinct fall, with lowest values between the twenty-fifth and thirty-fifth days followed by recovery with low normal average levels reached by about the fifty-fifth day. These changes in the total white count are due chiefly to variations in the number of neutrophils present. Abnormal giant neutrophils were

present early after irradiation. Other types of abnormal cells were seen in small numbers throughout the period of study.

The blood platelets may or may not have shown early increase above normal. The fluctuations during the first 2 weeks are not such as to be given any special significance. After that period there was a pronounced fall in the number of platelets with lowest values being reached between the twenty-fifth and thirtieth days. During the recovery phase there was a rebound to above normal values in some patients; the most pronounced elevation occurred in the patient who had had the highest radiation dose and who had shown the greatest thrombocytopenia around the twenty-fifth day.

Changes in the hemoglobin and hematocrit values were pronounced in only two patients. The greatest depression of red cell values was reached between the thirty-fifth and fortieth days; the lowest hematocrit value in one of these patients was about 30 and in the other about 25. These were the ones with the highest radiation dose and the most profound changes in the white cell and platelet levels. Recovery in hemoglobin and hematocrit values occurred rather gradually but there was a distinct reticulocyte increase between the thirty-fifth and fiftieth days, especially in one of the two highest-dose patients.

Bone-marrow studies were performed at frequent intervals. During the first 5 days after irradiation there was no striking drop in total cellularity but a slight relative decrease in red cell precursors. Giant granulocytes were seen and degenerative changes in megakaryocytes became prominent. On the ninth day after the irradiation the drop in total cellularity was pronounced and degenerative changes in the megakaryocytes were more prominent. Between the twenty-fourth and thirty-ninth days the marrows showed pronounced hypoplasia, and by this time there had been a profound decrease in granulocyte forms with a relative increase in red cell precursors and primitive and nondescript cells, some of which were difficult or impossible to identify. By the fifty-fourth day, the marrow showed active regeneration with hypercellularity.

Special studies done by FLIEDNER and his associates at the Brookhaven National Laboratory on this same group of patients, to be published elsewhere,[1] indicated a striking depression of the mitotic indices in the marrow, particularly on the fourth day after irradiation.

BIOCHEMICAL CHANGES

Many routine and specialized laboratory procedures were applied to the blood and urine of these patients, both as a matter of scientific interest and in an effort to evaluate their clinical condition as accurately as possible.

For the most part, the results were within the normal range of values or were not clearly ascribable to the effect of exposure. The taurine and β-aminoisobutyric acid excretion were, however, of greater interest. The preliminary results are summarized in Table 1.

Table 1. *Taurine and β-aminoisobutyric acid (BAIB) in the urine of accidentally irradiated humans*

Subject and interval after exposure	Taurine (mg/day)	BAIB (mg/day)*
A (365)† 4th day	273	54.8
A (365)† 16th day	146	
C (339) 1st day	204	38.4
E (236) 4th day	231	15.7
F (68) 4th day	109	13.6
I (0)‡	103	7.5

* To convert mg/day to μM/day, multiply the numbers in this column by 1000/103.1.

† The numbers in parenthesis are the doses in rads (γ + neutron).

‡ This was an ambulatory patient who had carcinoma of the lung but had not been exposed to any irradiation.

These results were obtained by applying the quantitative column chromatographic procedure of MOORE *et al.*[2] to aliquots of 24 hr urine collections.

The taurine excretion is elevated both 1 day and 4 days after exposure. By the sixteenth day taurine excretion had returned to the normal range in patient *A*. Taurine excretion is somewhat variable among normal individuals and is influenced by dietary intake. These are serious limitations if one desires to use the level

of taurine excretion as an indicator of the amount of irradiation to which the subject has been exposed. If one considers only the levels at 4 days, there appears to be a correlation with dosage. The small number of values available and the exception of patient *C* at 1 day make such a conclusion highly tenuous, however.

The β-aminoisobutyric acid excretion is elevated after irradiation in all of the subjects and appears to be correlated with dosage. This confirms RUBINI's[3] general conclusions from his study of these same patients. Other agents, causing DNA breakdown, can cause an elevated BAIB excretion; furthermore, some non-irradiated people habitually excrete 30 mg or more of this substance daily. An elevated excretion of this substance does not, therefore, necessarily indicate an exposure to irradiation.

DISCUSSION

This particular radiation accident provides much information on the changes following whole-body irradiation in the sublethal range. The fact that the patients were treated with great conservatism, without blood transfusions and with minimal antibiotics, seems to indicate that such conservative therapy is justifiable. Indeed, at the present state of our knowledge, such treatment may be preferable when the dose is believed to be definitely sublethal. One of the greatest problems is determining the dose fairly early after the irradiation. It is of value to pursue both physical measurements and estimates based upon clinical and laboratory observations of patients. A few examples of such studies can be cited here. General observation of the severity and persistence of the nausea and vomiting, and of the presence or absence of fever, is important. Significant temperature elevation in the first 48 hr would suggest a dose higher than that encountered in the present series. A pronounced fall in the lymphocytes suggests that the dose has been in the dangerous range, but the relationship is not clear between further increments in dose (in the range of 300 and 1000 rads, for example) and further depression in the lymphocytes. In the present series of cases the reticulocytes did not show any very pronounced fall during the first 3 or 4 days. It may be that by counting a large number of cells and obtaining a statistically accurate reticulocyte count, it would be possible to make an estimate of dosage. If there is an unequivocal fall in reticulocytes during the first 5 days, it is probable that the dose is higher than in the present series. Sodium activation studies, either based upon total body counts or blood radio-assays, have been shown to be of great value in estimating the neutron dosage. Early studies on β-aminoisobutyric acid and taurine may be established as valuable indices of radiation dosimetry.

Comparison of our experience with earlier reports of similar accidents and with the atom-bomb casualties in Japan, and the Marshallese Islanders makes it appear that whole-body irradiation produces rather predictable changes in man. The time sequence of these changes tends to remain fairly uniform for a given species over quite a large dose range, but there are very striking differences in the time sequence among different species. Small animals, in which much radiobiological research is done, show both the development and the recovery of the hematologic changes in a much shorter time than do human beings. The explanation of the delay in the development of maximal hematologic changes is quite complicated. Although the normal survival and life cycle of the different cell types are undoubtedly important, the timing of the changes cannot be explained entirely on this basis.

The study of the five patients in the Y-12 accident adds significantly to the information available on irradiation effects in human beings. More detailed information and analysis based on this experience will be presented.

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MAXIMUM LIKELIHOOD ANALYSIS OF IONIZING RADIATION INDUCED MORTALITY IN WHOLE BODY FRACTIONATED DOSE EXPERIMENTS

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Abstract—The type of experiment is considered in which a whole body dose of ionizing radiation is acutely administered to a test group of animals; after a specified period of time those still surviving are administered a second such dose. This procedure is repeatable on a number of such test groups using a different dose magnitude and intervals of separation for each group. Three exhaustive time intervals can be considered in which first interval deaths can be ascribed primarily to the first dose, those in the second to the additive effects of the second dose with the residual effect of the first dose, and those not dying until the third interval are considered survivors of the two doses. A mathematical model is derived for the analysis of such data that is a logical extension of the rationale used in the probit analysis of acute single dose data. Injury is postulated to be comprised of two additive components, one permanent and one recoverable. The recoverable injury decrements exponentially with time. Death occurs when the total injury exceeds the lethal threshold of an animal. The logarithm of the lethal thresholds of an animal at two different times is assumed to be bivariate normally distributed with a correlation depending upon the absolute magnitude of the difference between the two times. An iterative calculation procedure is presented for obtaining the maximum likelihood estimates of the relevant parameters and the variances and covariances of these estimates from experimental data of type described above.

THE simplest experiment to investigate the lethal effects of ionizing radiation is the single dose acute study. In this type of experiment a group of n_j animals is selected by a random process from the population under investigation and each subjected to a whole body dose of magnitude D_j delivered within a relatively short time interval. The proportion of the n_j animals dying within some specified time following the administration of the radiation is then observed. This procedure is repeated for a number of different dose magnitudes. The expected proportion killed at each dose level is a sigmoid shaped, monotonically increasing, function of the dose magnitude. The analysis of such data by use of probit and logit transforms has already been extensively discussed[1–3,9–12,15] in connection with the quantal response assay of various toxic materials and acute radiation damage studies.

A somewhat different type of experiment is that in which a group of animals is selected randomly from the population under investigation and these subjected to continuous constant low level radiation over a relatively long period of time until all of the members of the group have succumbed. The duration of life after onset of the irradiation is recorded for each animal. Thus, in experiments of this type, all the animals of the group eventually die but the time of death varies. A different group of animals is used for each different rate of dosage. It is found[23] that the mean life span under such circumstances is a monotonically decreasing function of the rate of dosage.

A third type is the fractionated dose experiment. In the simplest case of this type two groups, A and B, of animals would be drawn by random selection from the population under investigation. Group A would be given some dosage D. Group B would be given a dose D_1

and a time τ later the survivors of B administered a second dose D_2, such that $D_1 + D_2 = D$. Not many such studies have been done; however, in those of PATERSON *et al.*[21] and BLAIR[8] the proportion of animals dying for a given total dose in B was found to be less than that in A. The difference is an indication of the extent to which an animal recovers from a non-fatal dose in a time interval of length τ.

The analysis of such fractionated dose experiments has not been discussed with the thoroughness devoted to the single dose acute experiment. While the concept of radiation injury enters into discussions of both the "percent kill" of the single dose acute experiment and the life shortening of the chronic low level irradiation experiment, the operations by which it is measured in the two situations are not the same, nor is the relation between the two very clear. The fractionated dose experiment can be thought of as an intermediate form linking the acute and chronic irradiation experiments and is probably the most experimentally efficient method of estimating the recovery rate. It will be the purpose of this paper to discuss the analysis of a simple fractionated dose experiment.

Suppose k groups of animals are selected by some random process from the population under investigation. Designate the number allocated to the jth group as n_j where $j = 1, \ldots, k$. To each of the n_j animals of the jth group is administered a dose of magnitude D_j. At τ_j units of time after the administration of this first dose, those animals still surviving are each given a second dose of size D_j, i.e. the same size as the first dose. Designate the mean time lag, at which death following single dose irradiation with a dose D_j has been observed to occur, as W_j. Three time intervals shall be defined such that a death occurring in the first interval will most likely be due to the results of the first dose and one in the second most likely due to the resultant effects of the two doses. If an animal survives the acute effects of these two doses then it will be classed into the third interval. The precise meaning of these statements will become clearer later on in the discussion.

In keeping with the above requirements these three exhaustive time intervals are defined as follows:

designate $t = 0$ when first dose is given

$0 < t < t_1$ as first interval

$t_1 < t < t_2$ as second interval

$t_2 < t$ as third interval

with the convention that when

$W_j \leq \frac{1}{2}\tau_j$ then $t_1 = \tau_j$ and $t_2 = 2\tau_j$

$W_j > \frac{1}{2}\tau_j$ then $t_1 = W_j + \frac{1}{2}\tau_j$ and $t_2 = 2W_j + \tau_j$.

Let S_{1j} and S_{2j} denote the numbers of the jth group that die in the first and second intervals, respectively. Since the third interval is open ended all animals that do not die in the first and second intervals will eventually die in the third interval. These may be referred to alternately as "survivors" or third interval deaths. Thus

$$n_j = S_{1j} + S_{2j} + S_{3j} \qquad j = 1, \ldots, k$$

and the set of outcomes for all k groups will be

$$S_{11}, S_{21}, S_{31}, \ldots, S_{1j}, S_{2j}, S_{3j}, \ldots, S_{1k}, S_{2k}, S_{3k}$$

Let p_{1j}, p_{2j} and p_{3j} be the probability that an animal in the jth group will die in the first, second and third intervals, respectively.

Suppose one has a model concerning the radiation injury and rate of recovery that involves the parameters $\theta_1, \ldots, \theta_p$ $(p < 2k)$ such that if values are given for D_j, τ_j and $\theta_1, \ldots, \theta_p$ one could compute a unique set of values for p_{1j}, p_{2j} and p_{3j}. There will thus be three different functions, (two independent) involving D_j and τ_j as independent variables and $\theta_1, \ldots, \theta_p$ as parameters, whose values will be predictors for the expected proportion of animals of each group dying in the three time intervals. Then

$$p_{ij} = p_i(\theta_1, \ldots, \theta_p / D_j, \tau_j)$$

and

$$\sum_{i=1}^{3} p_{ij} = 1 \qquad i = 1, 2, 3;\ j = 1, \ldots, k$$

The pair of numbers D_j, τ_j associated with each group will obviously be experimental variables under the control of the experimenter and $\theta_1, \ldots, \theta_p$ will be characterizing quantities

of the population, to be estimated from the experimental outcome. It should be remarked that the pair of numbers D_j, τ_j will be unique for the jth group and will in fact constitute the defining property of those animals to be included in the jth group.

In addition to obtaining estimates of $\theta_1, \ldots, \theta_p$ one shall wish to obtain estimates of the variances and covariances of these estimates and to ascertain whether the experiment gives reason for doubting the validity of the model involving these parameters. To answer this latter question one will desire some kind of appropriate goodness of fit test.

MAXIMUM LIKELIHOOD ESTIMATION BY THE GARWOOD METHOD

With k different D_j, τ_j combinations and the probabilities $p_{ij} (i = 1, 2, 3;\ j = 1, \ldots, k)$, the probability of obtaining exactly the set of outcomes $S_{ij} (i = 1, 2, 3;\ j = 1, \ldots, k)$ will be

$$P = \prod_{j=1}^{k} \left(\frac{n_j}{S_{1j}!S_{2j}!S_{3j}!} p_{1j}^{S_{1j}} p_{2j}^{S_{2j}} p_{3j}^{S_{3j}} \right).$$

The liklihood, i.e. $\log P$, therefore is

$$L = \sum_{j=1}^{k} \log \frac{n_j}{S_{ij}!S_{2j}!S_{3j}!} + \sum_{j=1}^{3} \sum_{j=1}^{k} S_{ij} \log p_{ij}. \quad (1)$$

Maximum likelihood estimates of $\theta_1, \ldots, \theta_p$ can be computed by the iterative method outlined by GARWOOD (1941). The following discussion is based upon the GARWOOD procedure.

Define

$$L_h = \partial L / \partial \theta_h \qquad h = 1, \ldots, p \qquad (2)$$

their values being those obtained using the trial values $u_1, \ldots, u_p$ of $\theta_1, \ldots, \theta_p$.

For a given sample outcome $S_{11}, \ldots, S_{3k}$ the set of parameter values yielding the maximum value of L will be that set for which $L_h = 0$, $h = 1, \ldots, p$. Since the set of trial values will not usually be identical with this maximizing set, some or all of the $L_1, \ldots, L_p$ will usually be different from zero.

The expected values of the second derivatives of L with respect to the parameters shall be designated as L_{gh}, i.e.

$$L_{gh} = +E\left(\frac{\partial^2 L}{\partial \theta_g \, \partial \theta_h} \right) \qquad (3)$$

where

$$g = 1, \ldots, p$$
$$h = 1, \ldots, p$$

Let $U_1, \ldots, U_p$ be the maximum likelihood estimates of the θ's and $\delta u_1, \ldots, \delta u_p$ be the corrections on the trial estimates such that

$$\begin{aligned} U_1 &= u_1 + \delta u_1 \\ &\cdot \\ &\cdot \\ &\cdot \\ U_p &= u_p + \delta u_p \end{aligned} \qquad (4)$$

then

$$\sum_{g=1}^{p} L_{hg} \, \delta u_g = -L_h \qquad (5)$$

where $h = 1, \ldots, p$

will be the set of normal equations whose solutions will yield the first-order values of the corrections. If the parameter trial values chosen are sufficiently close to those actually yielding the maximal value of L then the first such set of corrections and corrected values of the parameters will be sufficient. If the initial set of trial values is chosen too far away from the maximum likelihood values then it may be necessary to use the corrected values obtained from the first calculation as the trial values for further iteration cycles.

Let

$$\Psi = [L_{hg}] \qquad (6)$$

denote the pxp symmetric matrix of the coefficients of the δu_g's in the set of normal equations (5), $|\Psi|$ denote the determinant of Ψ, Ψ^{-1} the inverse matrix of Ψ and $|\Psi_q|$ the value of the determinant obtained by replacing the qth column of Ψ by the column of terms $-L_h$ $(h = 1, \ldots, p)$. The solution for the corrections will then be

$$\delta u_q = \frac{|\Psi_q|}{|\Psi|} \qquad q = 1, \ldots, p. \qquad (7)$$

The inverse matrix Ψ^{-1} will be approximately the variance covariance matrix of the estimates $U_1, \ldots, U_p$ of the parameters $\theta_1, \ldots, \theta_p$.

The nature of the radiation damage theories with which we shall be concerned is such that the expressions yielding the values of the p_{ij}'s ($i = 1, \ldots, 3$; $j = 1, \ldots, k$) will be differentiable functions of $\theta_1, \ldots, \theta_p$. Differentiating (1) one obtains

$$\frac{\partial L}{\partial \theta_h} = \sum_{j=1}^{k} \left(\frac{S_{1j}}{p_{1j}} - \frac{S_{3j}}{p_{3j}} \right) \frac{\partial p_{1j}}{\partial \theta_h} + \sum_{j=1}^{k} \left(\frac{S_{2j}}{p_{2j}} - \frac{S_{3j}}{p_{3j}} \right) \frac{\partial p_{2j}}{\partial \theta_h} \qquad h = 1, \ldots, p. \quad (8)$$

Taking the second derivative one finds that

$$\begin{aligned} \frac{\partial^2 L}{\partial \theta_g \, \partial \theta_h} = & \sum_{j=1}^{k} \left(\frac{S_{1j}}{p_{1j}} - \frac{S_{3j}}{p_{3j}} \right) \frac{\partial^2 p_{1j}}{\partial \theta_g \, \partial \theta_h} \\ & + \sum_{j=1}^{k} \left(\frac{S_{2j}}{p_{2j}} - \frac{S_{3j}}{p_{3j}} \right) \frac{\partial^2 p_{2j}}{\partial \theta_g \, \partial \theta_h} \\ & - \sum_{j=1}^{k} \frac{S_{1j}}{p_{1j}^2} \frac{\partial p_{1j}}{\partial \theta_g} \frac{\partial p_{1j}}{\partial \theta_h} \\ & - \sum_{j=1}^{k} \frac{S_{3j}}{p_{3j}^2} \frac{\partial p_{1j}}{\partial \theta_g} \frac{\partial p_{1j}}{\partial \theta_h} \\ & - \sum_{j=1}^{k} \frac{S_{3j}}{p_{3j}^2} \frac{\partial p_{2j}}{\partial \theta_g} \frac{\partial p_{1j}}{\partial \theta_h} \\ & - \sum_{j=1}^{k} \frac{S_{2j}}{p_{2j}^2} \frac{\partial p_{2j}}{\partial \theta_g} \frac{\partial p_{2j}}{\partial \theta_h} \\ & - \sum_{j=1}^{k} \frac{S_{3j}}{p_{3j}^2} \frac{\partial p_{1j}}{\partial \theta_g} \frac{\partial p_{2j}}{\partial \theta_h} \\ & - \sum_{j=1}^{k} \frac{S_{3j}}{p_{3j}^2} \frac{\partial p_{2j}}{\partial \theta_g} \frac{\partial p_{2j}}{\partial \theta_h} \\ & g = 1, \ldots, p; \quad h = 1, \ldots, p. \end{aligned}$$

The expectations of S_{1j}, S_{2j} and S_{3j} will be $n_j p_{1j}$, $n_j p_{2j}$ and $n_j p_{3j}$, respectively. Since the expectation of a linear combination of stochastic variables is just the linear combination of the expectations, one obtains that

$$\begin{aligned} -E\left(\frac{\partial^2 L}{\partial \theta_g \, \partial \theta_h} \right) = & \sum_{j=1}^{k} n_j \left(\frac{1}{p_{1j}} + \frac{1}{p_{3j}} \right) \frac{\partial p_{1j}}{\partial \theta_g} \frac{\partial p_{1j}}{\partial \theta_h} \\ & + \sum_{j=1}^{k} n_j \left(\frac{1}{p_{2j}} + \frac{1}{p_{3j}} \right) \frac{\partial p_{2j}}{\partial \theta_g} \frac{\partial p_{2j}}{\partial \theta_h} \\ & + \sum_{j=1}^{k} \frac{n_j}{p_{3j}} \left(\frac{\partial p_{1j}}{\partial \theta_g} \frac{\partial p_{2j}}{\partial \theta_h} + \frac{\partial p_{1j}}{\partial \theta_h} \frac{\partial p_{2j}}{\partial \theta_g} \right). \quad (9) \end{aligned}$$

It is interesting to note in passing that the matrix elements L_{hg} can all be computed from a knowledge of the values of the probabilities p_{ij} and of their first derivatives $\partial p_{ij}/\partial \theta_g$. This is of considerable practical importance as will be seen later on when the explicit expressions involving the parameters $\theta_1, \ldots, \theta_p$ are being obtained for the theoretical model of interest.

CORRECTIONS FOR NORMAL DEATH RATE AND LAG VARIABILITY

The probabilities p_{1j} and p_{2j}, that an animal of the jth group will die in the first and second intervals, respectively, can each be viewed as the probability of the union of two separate events. These two events are (a) the animal dies in the interval of causes unrelated to radiation injury, (b) the animal dies in the interval from radiation injury.

Animals that have suffered fatal injury from radiation will exhibit variability in the time required for them actually to die. Because of this variability in the lag between the time of fatal injury and the time that death actually occurs there will be some animals fatally injured by the first dose not dying until the second interval. Similarly, there may be animals killed by the second dose dying in the first interval or surviving the second interval. It will be necessary to take these effects into account in some manner as well as the deaths due to "natural causes".

It has been suggested that animals dying from natural causes show an actuarial distribution in time adequately described by the solution of the differential equation

$$-\frac{1}{n}\frac{dn}{dt} = \alpha_0 e^{mt}$$

where n is the number remaining at age t and α_0 and m are constants.[13,18] In the case of non-senile animals and an interval of time Δt that is small in comparison to the mean life-span one can, however, to a satisfactory first approximation, for some purposes consider

$$\alpha_0 e^{m(t+\Delta t)} \doteq \alpha_0 e^{mt} \quad (10)$$

In the design under discussion the maximum interval under consideration will be twice the largest τ of interest. One will usually not be interested in values of τ more than several

times the value of the recovery time (this will be discussed more precisely later on) for experiments of the present design. For example, in the case of the rat the recovery half time has been estimated to be about 6 to 9 days.[14] Thus, even with values of τ five times the upper estimate of the recovery half time, the maximum interval under consideration would be not much larger than about 3 months or 10 per cent of the normal life span of the rat. With this in mind it shall be assumed that $-dn/dt = n\alpha$, where α is considered constant over Δt, gives an adequate description for the purpose of correcting for the natural death rate in the experimental situation under discussion.

To discuss the effect of the variability in lag time between radiation injury and death it will be assumed that the distribution of lag times for a given dose size can, for practical purposes, be considered normal with variance σ_w^2 and mean W. Since it is known that the mean lag is shorter for higher doses, W will be considered to be a function of the dose. Because negative lag times have no meaning and shall have a zero probability assigned to them, the density function will be normalized by multiplication by the factor $1/(1 - G(-W/\sigma_w))$ where G is used to denote the normal distribution function of mean zero and standard deviation unity.

Let p_{R1j} indicate the probability of fatal radiation injury being inflicted by the first dose. Let p_{R2j} be the joint probability of the first dose being non-fatal and the combined effects of the two doses being fatal. The possibility is not excluded that an animal may die of causes other than radiation injury even though it has received a fatal radiation injury. Define

$$g_1(t) = \frac{1}{\sigma_w\sqrt{(2\pi)}} \exp\left\{-\frac{(t-w)^2}{2\sigma_w^2}\right\} \tag{11}$$

$$g_2(t) = \frac{1}{\sigma_w\sqrt{(2\pi)}} \exp\left\{-\frac{(t-w-\tau)^2}{2\sigma_w^2}\right\} \tag{12}$$

$$G(t) = \frac{1}{\sqrt{(2\pi)}} \int_{-\infty}^{t} \exp\left(-\frac{t'^2}{2}\right) dt' \tag{13}$$

If the first dose is given to n_0 animals at $t = 0$, the differential equations describing the change in the number, n, with time will be

$$-\frac{dn}{dt} = \alpha n + \frac{n_0 e^{-\alpha t} p_{R1} g_1(t)}{1 - G(-W/\sigma_w)} \qquad \text{for } 0 < t < \tau$$

and

$$-\frac{dn}{dt} = \alpha n + \frac{n_0 e^{-\alpha t} p_{R1} g_1(t)}{\left(1 - G\left(\frac{-W}{\sigma_w}\right)\right)} + \frac{n_0 e^{-\alpha t} p_{R2}}{\left(1 - G\left(\frac{-W}{\sigma_w}\right)\right)} g_2(t)$$

for $t > \tau$. Integrating these with respect to time between the limits defining the three time intervals one obtains that

$$p_{1j} = (1 - e^{-\alpha t_{1j}}) + p_{R1j}C_{1j} + p_{R2j}C_{4j} \tag{14}$$

$$p_{2j} = (e^{-\alpha t_{1j}} - e^{-\alpha t_{2j}}) + (C_{2j} - C_{1j})p_{R1j} + (C_{3j} - C_{4j})p_{R2j} \tag{15}$$

$$p_{3j} = e^{-\alpha t_{2j}} - C_{2j}p_{R1j} - C_{3j}p_{R2j} \tag{16}$$

where

$$C_{1j} = \left(\frac{G\left(\frac{t_{1j} - W_j}{\sigma_w}\right) - G\left(\frac{-W_j}{\sigma_w}\right)}{1 - G\left(\frac{-W_j}{\sigma_w}\right)}\right) e^{-\alpha t_{1j}} \tag{17}$$

$$C_{2j} = \left(\frac{G\left(\frac{t_{2j} - W_j}{\sigma_w}\right) - G\left(\frac{-W_j}{\sigma_w}\right)}{1 - G\left(\frac{-W_j}{\sigma_w}\right)}\right) e^{-\alpha t_{2j}} \tag{18}$$

$$C_{3j} = \left(\frac{G\left(\frac{t_{2j} - W_j - \tau_j}{\sigma_w}\right) - G\left(\frac{-W_j}{\sigma_w}\right)}{1 - G\left(\frac{-W_j}{\sigma_w}\right)}\right) \times e^{-\alpha t_{2j}} \tag{19}$$

$$C_{4j} \begin{cases} = \left(\dfrac{G\left(\frac{t_{1j} - W_j - \tau_j}{\sigma_w}\right) - G\left(\frac{-W_j}{\sigma_w}\right)}{1 - G\left(\frac{-W}{\sigma_w}\right)}\right) \times e^{-\alpha t_{1j}} \\ \qquad \text{if } W_j < \tfrac{1}{2}\tau_j \\ = 0 \quad \text{if } W_j > \tfrac{1}{2}\tau_j. \end{cases} \tag{20}$$

The values of W and σ_w can be obtained from separate experimental data in which latency of death is measured as a function of dose for the

species and strain in question. A least squares regression, with W as the dependent variable, should be carried out for such data. The residual variance can then be used as the estimate of σ_w^2. The value of W_j to be used should be that corresponding to the dose D_j on the regression curve. With the appropriate value of W_j known for each group and the value of τ_j known one can compute the values of $C_{1j}, \ldots, C_{4j}$ and set the limits on the intervals for $j = 1, \ldots, k$. The values of W_j and σ_w, it should be noted, are not included in the parameters $\theta_1, \ldots, \theta_p$ since they are obtained from experimental data other than the set under analysis. This is an approximation although, in the writer's opinion, a justifiable one. The unirradiated death rate α will, however, be included among the $\theta_1, \ldots, \theta_p$. To avoid confusion in the notation let us arbitrarily agree to identify α with the generalized parameter θ_p.

With this understanding, differentiation of (14) and (15) gives for $g = p$

$$\frac{\partial p_{1j}}{\partial \theta_g} = t_{1j}e^{-\alpha t_{1j}} - t_{1j}C_{1j}p_{R1j} - t_{1j}C_{4j}p_{R2j} \quad (21)$$

$$\frac{\partial p_{2j}}{\partial \theta_g} = (t_{2j}e^{-\alpha t_{2j}} - t_{1j}e^{-\alpha t_{1j}}) + (t_{1j}C_{1j} - t_{2j}C_{2j})p_{R1j} + (t_{1j}C_{4j} - t_{2j}C_{3j})p_{R2j} \quad (22)$$

and for $g = 1, \ldots, 1 - p$

$$\frac{\partial p_{1j}}{\partial \theta_g} = C_{1j}\frac{\partial p_{R1j}}{\partial \theta_g} + C_{4j}\frac{\partial p_{R2j}}{\partial \theta_g} \quad (23)$$

$$\frac{\partial p_{2j}}{\partial \theta_g} = (C_{2j} - C_{1j})\frac{\partial p_{R1j}}{\partial \theta_g} + (C_{3j} - C_{4j})\frac{\partial p_{R2j}}{\partial \theta_g}. \quad (24)$$

THE MECHANISM OF THE RADIATION INDUCED LETHAL PROCESS

To obtain the expressions for computing the probabilities p_{R2j} and $\partial p_{R2j}/\partial \theta_g$ $(g = 1, \ldots, p - 1)$ in terms of the dosage, the interval of separation of the dosages, and the trial values of $\theta_1, \ldots, \theta_p$, it is necessary to postulate a mechanism for death produced by whole body irradiation.

The assumed mechanism will, aside from a few simple but important innovations, resemble those proposed earlier by SACHER,[23] BLAIR[6,7] and SMITH[24] and reviewed by BERLIN and DIMAGGIO[4]. It will be assumed that the ionizing radiation produces some kind of cellular injury, a portion of which the organism can repair and a portion that is irreparable. If this injury exceeds some threshold (or as SACHER[23] prefers to call it, a "lethal bound") value of the animal then the animal dies. If this threshold is not exceeded the animal does not die (or at least not from radiation injury). The dimensions of both the injury and the threshold will be those of the roentgen unit. This paper will be exclusively concerned with kill data of the type mentioned earlier, i.e. the set of S_{ij} values, where $i = 1, 2, 3$ and $j = 1, \ldots, k$. A discussion of the relationship between this type of data and the shortening of life expectancy observed in chronic irradiation is given in another paper.[5]

Let $\gamma(t)$ be the rate of whole body dosage, measured in roentgens per unit time, at time t. The parameter A is a dimensionless constant, greater than zero and less than one, that shall be interpreted as the irreparable fraction of the injury. The parameter B will be the time constant indicating the rate of recovery and will have the dimensions of reciprocal time. The amount of reparable and irreparable injury shall be designated as r_R and r_I, respectively and both expressed in units of roentgens. The total injury r at any time will be assumed to be the sum of r_R and r_I at that time. It is assumed that

$$\frac{dr_I}{dt} = A\gamma(t) \quad (25)$$

$$\frac{dr_R}{dt} = (1 - A)\gamma(t) - Br_R \quad (26)$$

and that $r_R = r_I = 0$ at $t = 0$.

Integrating these two equations, the total injury r at t will be

$$r = A\int_0^t \gamma(t')\,dt' + (1 - A)e^{-Bt} \times \int_0^t \gamma(t')e^{Bt'}\,dt' \quad (27)$$

In the particular case of interest, in which two doses of size D each are administered τ time units apart and the time in which each of the doses is delivered is small in comparison to $1/B$, the maximum value of r at the first dose will be

$$r_1 = D \tag{28}$$

and at the second dose will be

$$r_2 = (1 + A + (1 - A)e^{-B\tau})D. \tag{29}$$

Since equations (25) and (26) are formally identical with those of SMITH[24] equations (28) and (29) are the same as those yielded by the Smith theory for this situation. At this point the present treatment and the Smith theory depart from one another.

It is frequently found for single doses of a toxic agent that the logarithm of the just lethal dose for each animal is normally distributed among the animals of the test population. The thresholds of an animal in the two trials will therefore be tentatively assumed to have a joint probability distribution that is a bivariate normally distributed function of $\log r_1$ and $\log r_2$. The correlation of the two thresholds will in general be a function of their temporal separation τ. The variance of the threshold at the time of the second dose will be assumed to be the same as that at the time of the first dose. If σ^2 is the variance, μ the mean and ρ_j the correlation of the logarithm of the two thresholds then

$$p_{R1j} = \int_{-\infty}^{h_j}\int_{-\infty}^{+\infty} \frac{1}{2\pi\sqrt{(1-\rho_j{}^2)}} \times \exp\left\{-\frac{1}{2(1-\rho_j{}^2)} \times (x_1{}^2 + x_2{}^2 - 2\rho_j x_1 x_2)\right\} dx_1\, dx_2 \tag{30}$$

$$p_{R2j} = \int_{h_j}^{\infty}\int_{-\infty}^{k_j} \frac{1}{2\pi\sqrt{(1-\rho_j{}^2)}} \times \exp\left\{-\frac{1}{2(1-\rho_j{}^2)} \times (x_1{}^2 + x_2{}^2 - 2\rho_j x_1 x_2)\right\} dx_1\, dx_2 \tag{31}$$

$$p_{R3j} = \int_{h_j}^{\infty}\int_{k_j}^{\infty} \frac{1}{2\pi\sqrt{(1-\rho_j{}^2)}} \times \exp\left\{-\frac{1}{2(1-\rho_j{}^2)} \times (x_1{}^2 + x_2{}^2 - 2\rho_j x_1 x_2)\right\} dx_1\, dx_2 \tag{32}$$

where

$$h_j = \frac{1}{\sigma}(\log r_{1j} - \mu) = \frac{1}{\sigma}\log D_j - \frac{\mu}{\sigma} \tag{33}$$

$$k_j = \frac{1}{\sigma}(\log r_{2j} - \mu) = \frac{1}{\sigma}\log D_j + \frac{1}{\sigma}\log(1 + A + (1 - A)e^{-B\tau_j}) - \frac{\mu}{\sigma}. \tag{34}$$

OWEN[20] has derived a useful procedure that shall be employed for the evaluation of these bivariate normal probability integrals.

Define

$$M(h, k;\ \rho) = \frac{1}{2\pi\sqrt{(1-\rho^2)}} \int_h^{\infty}\int_k^{\infty} \exp\left(-\frac{1}{2}\frac{x^2 + y^2 - 2\rho xy}{1-\rho^2}\right) dx\, dy \tag{35}$$

$$T(h, a) = \frac{1}{2\pi}\int_0^a \frac{\exp(-\frac{1}{2}h^2(1 + x^2))}{1 + x^2}\, dx. \tag{36}$$

OWEN has demonstrated the following relationships to be true

$$M(h, k;\ \rho) = 1 - \tfrac{1}{2}G(h) - \tfrac{1}{2}G(k) - T\left(h, \frac{k - \rho h}{h\sqrt{(1-\rho^2)}}\right) - T\left(k, \frac{h - \rho k}{k\sqrt{(1-\rho^2)}}\right)$$

if $hk > 0$, or if $hk = 0$ and h or $k > 0$, or if both $= 0$, (37)

$$M(h, k;\ \rho) = \tfrac{1}{2} - \tfrac{1}{2}G(h) - \tfrac{1}{2}G(k) - T\left(h, \frac{k - h}{h\sqrt{(1-\rho^2)}}\right) - T\left(k, \frac{h - k}{k\sqrt{(1-\rho^2)}}\right)$$

if $hk < 0$, or if $hk = 0$ and h or $k < 0$

and has tabulated the values of $T(h, a)$ as a function of h and a.

Define a function $I(h, k)$ such that

$$\left.\begin{aligned} &I = 0 \text{ if } hk > 0, \text{ or if } hk = 0 \text{ and } h \text{ or } k > 0, \text{ or if both } = 0 \\ &I = 1 \text{ if } hk < 0, \text{ or if } hk = 0 \text{ and } h \text{ or } k < 0. \end{aligned}\right\} \tag{38}$$

Making use of relations (30), (31), (32) and (37) and definitions (35), (36) and (38), one can write the expressions for p_{R1j}, p_{R2j} and p_{R3j} in terms of functions whose values have been tabulated; thus one obtains

$$p_{R1j} = G(h_j) \tag{39}$$

$$\begin{aligned} p_{R2j} &= \tfrac{1}{2}G(k_j) - \tfrac{1}{2}G(h_j) \\ &\quad + T\left(h_j, \frac{k_j - \rho_j h_j}{h_j\sqrt{(1-\rho_j^2)}}\right) \\ &\quad + T\left(k_j, \frac{h_j - \rho_j k_j}{k_j\sqrt{1-\rho_j^2}}\right) + \tfrac{1}{2}I(h_j, k_j) \end{aligned} \tag{40}$$

$$\begin{aligned} p_{R3j} &= 1 - \tfrac{1}{2}G(h_j) - \tfrac{1}{2}G(k_j) \\ &\quad - T\left(h_j, \frac{k_j - \rho_j h_j}{h_j\sqrt{(1-\rho_j^2)}}\right) \\ &\quad - T\left(k_j, \frac{h_j - \rho_j k_j}{k_j\sqrt{(1-\rho_j^2)}}\right) - \tfrac{1}{2}I(h_j, k_j). \end{aligned} \tag{41}$$

It will be noted that, although $I(h, k)$ and $T\left(h, \frac{k-\rho h}{h\sqrt{(1-\rho^2)}}\right) + T\left(k, \frac{h-\rho h}{k\sqrt{(1-\rho^2)}}\right)$ are separately discontinuous at $h = 0$ and at $k = 0$, p_{R2} and p_{R3} are everywhere continuous and their first derivatives with respect to h and k defined. This is important in that it will mean that the first order expansions in terms of the corrections on the parameter trial values will remain valid even though the trial values of h and k may be such that $kh > 0$ and the corrected values such that $hk < 0$, or vice versa.

From equations (8) and (9) it was seen that in order to compute the values of $L_{h\theta}$ and L_h it is necessary to obtain suitable expressions for computing trial values of $\partial p_{R1j}/\partial\theta_g$ and $\partial p_{R2j}/\partial\theta_g$.

It can be demonstrated that

$$\frac{\partial T(h, a)}{\partial h} = (G(ha) - \tfrac{1}{2})g(h) \tag{42}$$

$$\frac{\partial T(h, a)}{\partial a} = \frac{g\{h\sqrt{(1+a^2)}\}}{(1+a^2)\sqrt{2\pi}} \tag{43}$$

where $g(x) = \dfrac{1}{\sqrt{2\pi}} \exp\left(-x^2/2\right)$

Define

$$\psi_2(h, a) = \partial T(h, a)/\partial h \tag{44}$$

$$\psi_3(h, a) = \partial T(h, a)/\partial a \tag{45}$$

$$a_1 = \frac{k - \rho h}{h\sqrt{(1-\rho^2)}}, \quad a_2 = \frac{h - \rho k}{k\sqrt{(1-\rho^2)}}. \tag{46}$$

To facilitate calculation the values of $\psi_2(h, a)$ and $\psi_3(h, a)$ have been computed from equations (42) and (43) and presented in tabular form in the Appendix.

With relations (42) and (43) and definitions (44), (45) and (46) expression (40) can be differentiated with respect to h and k to obtain that

$$\begin{aligned} \frac{\partial p_{R2j}}{\partial h_j} &= -\tfrac{1}{2}g(h_j) + \psi_2(h_1, a_{1j}) \\ &\quad - \frac{k_j}{k_j^2\sqrt{(1-\rho_j^2)}}\,\psi_3(h_j, a_{1j}) \\ &\quad + \frac{1}{k_j\sqrt{(1-\rho_j^2)}}\,\psi_3(k_j, a_{2j}), \end{aligned} \tag{47}$$

$$\begin{aligned} \frac{\partial p_{R2j}}{\partial k_j} &= \tfrac{1}{2}g(k_j) + \psi_2(k_j, a_{2j}) \\ &\quad - \frac{h_j}{k_j^2\sqrt{(1-\rho_j^2)}}\,\psi_3(k_j, a_{2j}) \\ &\quad + \frac{1}{h_j\sqrt{(1-\rho_j^2)}}\,\psi_3(h_j, a_{1j}). \end{aligned} \tag{48}$$

It will be useful at this point in the discussion to introduce a simple mathematical device for avoiding ambiguity in the notation. Let T_z, $z = 1, \ldots, p-5$, be the complete set of different experiment values of τ_j used in the experiment. That is to say no two values of T_z are the same and there is no value of the set of $\tau_j, j = 1, \ldots, k$, that does not occur in the set T_z. To each value of T_z there will correspond a single value of ρ_z. This set of ρ_z's, $z = 1, \ldots, p-5$, will be the $p-5$ values of the correlation to be estimated. Define a characteristic function $Q(z, j)$ such that if

$$\begin{aligned} T_z = \tau_j \quad &\text{then} \quad Q(z, j) = 1 \\ T_z \neq \tau_j \quad &\text{then} \quad Q(z, j) = 0. \end{aligned} \tag{49}$$

Clearly then

$$\rho_j = \sum_{z=1}^{p-5} Q(z, j)\rho_z \tag{50}$$

for any $j = 1, \ldots, k$. As a further safeguard against confusion let us agree to identify $\theta_1, \ldots, \theta_{p-5}$ with the ρ_z's and $\theta_{p-4}, \ldots, \theta_{p-1}$ with μ, $1/\sigma$, B and A. θ_p has, of course, already been identified with α.

Differentiating (39) and (40) with respect to the parameters and substituting into equations (23) and (24) one can write the required expressions for $\partial p_{ij}/\partial\theta_g$ ($i = 1, ; j = 1, \ldots, k$; $g = 1, \ldots, p-1$).

$$\frac{\partial p_{1j}}{\partial \rho_z} = -Q(z,j)\frac{C_{4j}}{h_j k_j(1-\rho_j^2)} \{k_j^2 a_{2j}\psi_3(h_j, a_{1j}) + h_j^2 a_{1j}\psi_3(k_j, a_{2j})\} \quad (51)$$

$$\frac{\partial p_{2j}}{\partial \rho_z} = -Q(z,j)\frac{(C_{3j}-C_{4j})}{h_j k_j(1-\rho_j^2)} \{k_j^2 a_{2j}\psi_3(h_j, a_{1j}) + h_j^2 a_{1j}\psi_3(k_j, a_{2j})\} \quad (52)$$

where $z = 1, \ldots, p-5$;

$$\frac{\partial p_{1j}}{\partial \mu} = -\frac{C_{1j}}{\sigma}g(h_j) - \frac{C_{4j}}{\sigma}\left(\frac{\partial p_{R2j}}{\partial h_j} + \frac{\partial p_{R2j}}{\partial k_j}\right) \quad (53)$$

$$\frac{\partial p_{2j}}{\partial \mu} = -\frac{1}{\sigma}(C_{2j}-C_{1j})g(h_j) - \frac{1}{\sigma}(C_{3j}-C_{4j}) \times \left(\frac{\partial p_{R2j}}{\partial h_j} + \frac{\partial p_{R2j}}{\partial k_j}\right) \quad (54)$$

$$\frac{\partial p_{1j}}{\partial(1/\sigma)} = C_{1j}(\ln D_j - \mu)g(h_j) + C_{4j}(\ln D_j - \mu) \times \left(\frac{\partial p_{R2j}}{\partial h_j} + \frac{\partial p_{R2j}}{\partial k_j}\right) + C_{4j}\left(\frac{\partial p_{R2j}}{\partial k_j}\right) \times \ln(1 + A + (1-A)e^{-B\tau_j}) \quad (55)$$

$$\frac{\partial p_{2j}}{\partial(1/\sigma)} = (C_{2j}-C_{1j})g(h_j)(\ln D_j - \mu) + (C_{3j}-C_{4j})(\ln D_j - \mu)\left(\frac{\partial p_{R2j}}{\partial h_j} + \frac{\partial p_{R2j}}{\partial k_j}\right) + (C_{3j}-C_{4j})\left(\frac{\partial p_{R2j}}{\partial k_j}\right) \times \ln(1 + A + (1-A)e^{-B\tau_j}) \quad (56)$$

$$\frac{\partial p_{1j}}{\partial A} = \frac{C_{4j}}{\sigma}\frac{(1-e^{-B\tau_j})}{(1+A+(1-A)e^{-B\tau_j})}\left(\frac{\partial p_{R2j}}{\partial k_j}\right) \quad (57)$$

$$\frac{\partial p_{2j}}{\partial A} = \frac{(C_{3j}-C_{4j})}{\sigma}\frac{(1-e^{-B\tau_j})}{(1+A'+(1-A')e^{-B\tau_j})} \times \left(\frac{\partial p_{R2j}}{\partial k_j}\right) \quad (58)$$

$$\frac{\partial p_{1j}}{\partial B} = -\frac{C_{4j}}{\sigma}\frac{\tau_j(1-A)e^{-B\tau_j}}{(1+A(1-A)e^{-B\tau_j})}\left(\frac{\partial p_{2j}}{\partial k_j}\right) \quad (59)$$

$$\frac{\partial p_{2j}}{\partial B} = -\frac{(C_{3j}-C_{4j})}{\sigma}\frac{\tau_j(1-A)e^{-B\tau_j}}{(1+A+(1-A)e^{-B\tau_j})} \times \left(\frac{\partial p_{R2j}}{\partial k_j}\right). \quad (60)$$

In expressions (53) through (60) in which $\partial p_{R2j}/\partial h_j$ and $\partial p_{R2j}/\partial k_j$ appear, the values computed from equations (47) and (40) should be used. The probability values p_{R1j}, p_{R2j} and p_{R3j} needed for substitution into expressions (14), (15) and (16) to obtain the values of p_{1j}, p_{2j} and p_{3j} and needed for substitution into (21) and (22) to obtain values of $\partial p_{1j}/\partial\alpha$ and $\partial p_{2j}/\partial\alpha$, can be computed from (39), (40) and (41). *It is to be understood, of course, that the trial values of ρ_z, μ, $1/\sigma$, B, A and α are to be used wherever these quantities appear in the computational expressions. If a second iteration is needed the parameter estimates to be used will be the corrected values from the preceding computation cycle.*

To recapitulate briefly, the process by which the maximum likelihood estimates of the parameters ρ_z, μ, $1/\sigma$, B, A and α (if there are $p-5$ different values of τ_j used then $z = 1, \ldots, p-5$) are obtained is as follows. Let us suppose, for purposes of illustration, that three different dose spacings τ_j have been used and there have been six different dose magnitudes D_j employed for each different τ_j value in addition to an unirradiated control group. There will therefore be 19 different pairs of values D_j, τ_j. The number of groups, k, of animals will thus be 19. Since in each group there are three things (death in the first, second or third interval) that can happen to an animal there will be 57 categories in which frequencies will be observed, 38 of which are independent. To account for the frequencies in these 38 independent categories one has 8 parameters. There will thus be 30 residual degrees of freedom for testing the radiation damage hypothesis. The values of W_j, necessary for determining the boundaries of the three intervals, are determined from separate experimental data in which the lag between fatal dosage and death is observed as a function of the dosage. The variance σ_w^2 of the lag for a particular dose value is obtained

from the same set of separate experimental data. The parameters $\theta_1, \ldots, \theta_p$ are estimated as follows:

A set of trial values $u_1, \ldots, u_p$ $(p = 8)$ is chosen for the parameters ρ_1, ρ_2, ρ_3, μ, $1/\sigma$, B, A and α. These should be the best preliminary estimates one has at one's disposal. With the trial value of α and the *a priori* (from other data) values of W_j and σ_w, compute the values of C_{1j}, C_{2j}, C_{3j} and C_{4j} from equations (17) through (20). From equations (33) and (34), the parameter trial values and the known values of D_j and τ_j, compute h_j and k_j for each of the 19 groups. From equations (39), (40) and (41) compute the trial values of p_{R1j}, p_{R2j} and p_{R3j} with the help of the tabulated values of $T(h, a)$ of OWEN (1957). Compute $\partial p_{R2j}/\partial h_j$ and $\partial p_{R2j}/\partial k_j$ from equations (47) and (48), using the tables for $\psi_2(h, a)$ and $\psi_3(h, a)$ given in the appendix of the present paper. With the values of $\partial p_{R2j}/\partial h_j$, $\partial p_{R2j}/\partial k_j$, C_{1j}, C_{2j}, C_{3j} and C_{4j} that have been computed and the trial values of the parameters, use equations (51) through (60) to compute

$$\partial p_{ij}/\partial \theta_g (1 = 1, 2;\ j = 1, \ldots, k;\ g = 1, \ldots, p - 1).$$

Use equations (14), (15) and (16) to compute the trial values of p_{1j}, p_{2j} and p_{3j} with the trial values of p_{R1j}, p_{R2j} and p_{R3j} already computed. Using the computed trial values of p_{1j}, p_{2j}, p_{3j}, $\partial p_{1j}/\partial \theta_g$ and $\partial p_{2j}/\partial \theta_g$, compute the values of the L_h from equation (8) and L_{gh} from equation (9) for $g = 1, \ldots, 8$; $h = 1, \ldots, 8$.

Equation (7) is then used to compute the corrections $\delta u_q (q = 1, \ldots, 8)$ to be added to the trial values $u_1, \ldots, u_8$ to obtain the corrected values $U_1, \ldots, U_8$ of the parameters. The inverse matrix Ψ^{-1} is then computed and the variances and covariances of the parameter estimates obtained.

In view of the fact that the computational procedure, although straightforward, is somewhat lengthy, it is recommended that it be programmed for electronic digital computation. Since a considerable proportion of the establishments concerned with research on radiation damage have access to such computing facilities, this should not present too severe a handicap in the application of the theory.

Two questions in respect to the model and the procedure have not yet been answered.

A question may be raised concerning the estimation of the normal death rate parameter, α. In obtaining the values of W_j and σ_w from auxiliary experimental data is it not necessary to correct for the normal death in some manner and by so doing may it not be possible that one is biasing the estimate of α in the paired dose experiment? In a strictly logical sense this point must be conceded; however, it should be pointed out that one would never conduct such a paired dose experiment as a way of estimating α. It enters into these experiments only as a control parameter to correct for the normal death rate which will not be large for the intervals of time of interest. Hence, any bias introduced into the parameters of interest by this procedure will be second-order magnitude effects and, for practical purposes, non-existent.

TEST OF AGREEMENT OF THE MODEL WITH THE DATA AND RELATED MATTERS

Several questions in respect to the model and the procedure have not yet been discussed. It is obviously necessary to know when the sequence of iterations can be expected to converge to an extremal value of L, whether or not that extremal value is a maximum, and at what iteration step the computed parameter values are sufficiently close to the extremal point values that no further correction is necessary. Finally, there is the problem of ascertaining whether there is reason to suspect that the assumed model, with the maximum likelihood estimates of the parameters, does not describe the data, i.e. the problem of "goodness of fit". In the subsequent discussion there will be some reference to the matrix Ψ. Most of these remarks are rigorously applicable only to the matrix whose elements are $\partial^2 L/\partial \theta_h \partial \theta_g$; however, it will be assumed that this matrix is adequately approximated by its expectation Ψ.

Although the set of parameter values yielding the maximum of L is the one desired it should be noted that sets of parameter values coinciding with minima or "saddle points" of L will also satisfy the condition $L_1 = \ldots = L_p = 0$. If a

set of parameter values for which $L_h = 0$ for $h = 1, \ldots, p$ yields a matrix Ψ that is

(1) Positive definite,* L is a minimum.
(2) Negative definite,* L is a maximum.
(3) Singular, the test fails.
(4) None of the above, L is at a saddle point.

Furthermore, there will exist a neighborhood of parameter values about the maximum point such that Ψ is negative definite for every set of parameter values in that neighborhood. This has relevance to the appropriate choice of an iteration sequence.

Denote the change in L associated with the parameter corrections computed in an iteration as ΔL, the set of values of the first derivatives $L_1, \ldots, L_p$ written in form of a column vector as $\mathbf{L}$ and the same set in the form of a row vector as $\mathbf{L}'$. To a linear approximation $\Delta L = -\mathbf{L}'\Psi^{-1}\mathbf{L}$. Hence ΔL can be expected to be positive if for every non zero $\mathbf{L}$, $\mathbf{L}'\Psi^{-1}\mathbf{L} < 0$. This is equivalent to the assertion that Ψ^{-1}, and hence Ψ, be negative definite. In order to have assurance that the sequence of iterations will converge upon a maximum point of L the following rule is suggested by the preceding considerations. Use as trial values, $u_1, \ldots, u_p$, only those sets yielding a negative definite matrix Ψ. If the set of parameter values obtained at any point in the iteration is such that Ψ is not negative definite, then that iteration sequence should be dropped and a new set of trial values chosen from which a new iteration sequence is initiated.

If the iteration sequence is convergent there will be some iteration cycle yielding a corrected set of parameter estimates sufficiently close to the set for which $L_1 = , \ldots = L_p = 0$ that no further corrections are necessary. As an estimate of the sampling variance to be anticipitated in the value of the derivative L_g one can use the expression

$$\text{Var}\,(L_g) = \Psi_g'\Psi^{-1}\Psi_g \qquad (61)$$

where Ψ_g and Ψ_g' are, respectively, the column and row vectors comprised of the elements $L_{1g}, L_{2g}, \ldots, L_{pg}$. The iteration should be terminated when the biggest value of $L_g^2/\text{Var}\,(L_g)$ is small in comparison to one.

When the convergent iteration has been terminated one will wish to ascertain whether the maximized value of the likelihood under the constraints imposed by the model is significantly different from the maximum value of the likelihood in the absence of those constraints, i.e. a test of "goodness of fit". Let $L^{(2k)}$ denote the value of the likelihood function (1) with the P_{ij} set equal to S_{ij}/n_j for $i = 1, 2, 3$ and $j = 1, \ldots, k$. Let $L^{(p)}$ designate the maximized value of the likelihood function under the constraints imposed by the model. For large sample sizes and adequate randomization one can use $\chi^2 = 2(L^{(2k)} - L^{(p)})$ with $2k - p$ degrees of freedom as the test statistic for testing for "goodness of fit" of the theory with the data. From expression (1) and the definitions of $L^{(2k)}$ and $L^{(p)}$ one finds that

$$\chi^2 = 2\sum_{i=1}^{3}\sum_{j=1}^{k} S_{ij}\log\left(\frac{S_{ij}}{n_j P_{ij}^{\star}}\right) \qquad (62)$$

where $P_{1j}^{\star}$, $P_{2j}^{\star}$ and $P_{3j}^{\star}$ are the values of p_{1j}, p_{2j} and p_{3j} obtained from equations (14), (15), (16), (39), (40) and (41) using the corrected estimates of the parameters ρ_z, μ, $1/\sigma$, A, B and α.

Thus, if the n_j are large and the discrepancies of the observed from the predicted number falling in each class are merely due to random sampling errors, expression (62) reduces to the conventional form

$$\chi^2 = \sum_{i=1}^{3}\sum_{j=1}^{k}\frac{(S_{ij} - n_j P_{ij}^{\star})^2}{n_j P_{ij}^{\star}} \qquad (63)$$

with $2k - p$ degrees of freedom, which will be recognized as the conventional χ^2-test for a hypothesis concerning the frequency of occurrence in a number of different classes.

SOME COMMENTS ON THE MEANING OF ρ

An important parameter contained in the present theoretical model not present in the Blair or Smith interpretation of fractionated dose effects is the threshold correlation ρ_z. Up to this point in the discussion no mention has been made of its interpretation although the procedure for its estimation has been give

* For a discussion of the definiteness of a matrix and the practical way of diagnosing it the reader is referred to Rao,[22] pp. 19–21.

One can look upon the lethal threshold of an animal as a time series. In the case of n_j animals there will be n_j such time series. In the present theory it has been implicitly assumed that the lethal threshold can be viewed as a stationary stochastic time series over intervals of the order of magnitude of T_z. An obvious generalization to the case in which the threshold is not a stationary time series would be to make μ a function of time.

One may be inclined to feel there is no operational significance in conceiving of an individual as having more than one lethal threshold value, and therefore none in speaking of the correlation between successive values of such a threshold. Suppose, however, that each animal had some unique value for its lethal threshold, i.e. that it was the same for a given animal at all times, and the following experiment was performed. The single dose lethal threshold is determined for a large number of animals and a conventional sigmoid shaped mortality

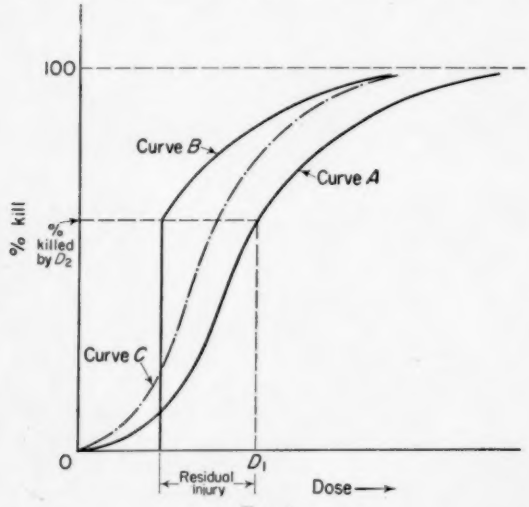

FIG. 1.

curve is obtained as shown in curve A of Fig. 1. Another large group of animals, drawn from the same parent population, is administered a dose D_1. After a period of time the survivors of the dose D_1 are administered a second dose D_2 such that one determines the percentage kill as a function of the magnitude of D_2. If the injury is considered, as in the present treatment, to be a deterministic quantity and the lethal threshold of an individual is single valued then one would find, among the animals receiving the second dosage, that none would be killed unless D_2 + residual injury is greater than D_1 and, in the dose interval where it is, the curve would parallel the expected mortality curve A for the single dose group. Thus, under such circumstances, one would find a curve of the sort depicted by curve B of Fig. 1. If, on the other hand, the threshold of an individual is not unique but has two different values at the two dose times then one would expect to find a curve more like C of Fig. 1. Since such an experiment will operationally detect whether the animals in a population each have unique or multiple valued lethal thresholds, it is clearly meaningful to speak of a temporal sequence of lethal threshold values in a single animal and of any of the statistical parameters characterizing such a sequence.

Although ρ_z is related to the autocorrelation, a_z, of the threshold of an animal when the lag time is T_z, these quantities are not identical. Let m denote the average threshold of a given animal and σ_m^2 the variance of m among the different animals of the population. Let σ_s^2 be the variance of the threshold in the temporal sequence of a given individual. The relation between a_z and ρ_z is

$$\rho_z^2 = (a_z^2 + \sigma_m^2/\sigma_s^2)/(1 + \sigma_m^2/\sigma_s^2).$$

If the intra-animal threshold variability is large in comparison to the inter-animal variability in m then σ_m^2/σ_s^2 will be small and ρ^2 will be approximately equal to a^2. Cyclical variations, e.g. seasonal and oestrous cycles, in the threshold may occur, with a period Y tending to be similar for the various animals of the population in which case ρ_z^2 would tend to be a maximum at that z for which $T_z \doteq Y$. If σ_m^2/σ_s^2 is large then this autocorrelation effect will tend to be masked. On the other hand, a large σ_m^2/σ_s^2 means that selection effects are playing an appreciable role. Importance of the autocorrelation effect means that the animal's relative survival chances are largely determined by the phase of its physiological cycle at which it happens to be dosed. Importance of the selection effect means that an

animal's relative survival changes are largely determined by its intrinsic radiation resistance.

Two important questions may thus be asked of the correlation estimates ρ. First, are the values of ρ_z the same for the various values of T_z, i.e. can the difference among the various ρ_z estimates be accounted for by random sampling variation? Secondly, are the values of ${\rho_z}^2 > 0$? Space does not permit detailed description of the computation for these two tests; however, the manner in which they can be carried out will be briefly outlined.

In addition to the maximum likelihood estimates of the correlations one has estimates of the variances and covariances of these estimates. The strategy to be suggested involves transformation of the ρ's to a set of approximately normally distributed variables whose covariances are all zero then using χ^2 as a test statistic. Making use of the suggestion of FISHER[16] (p. 197) the correlations $\rho_1, \ldots, \rho_b$ are transformed to the variables $\omega_1, \ldots, \omega_b$ where

$$\omega_j = \tfrac{1}{2} \log (1 + \rho_j) - \tfrac{1}{2} \log (1 - \rho_j)$$

$$\text{Var } (\omega_j) = \frac{1}{2(1 - {\rho_j}^2)^2} \text{Var } (\rho_j) \qquad (64)$$

$$\text{Cov } (\omega_i, \omega_j) = \frac{1}{2(1 - {\rho_i}^2)(1 - {\rho_j}^2)} \text{Cov } (\rho_i, \rho_j).$$

The $\omega_1, \ldots, \omega_b$ are then transformed to a set of variables $v_1, \ldots, v_b$ such that Cov $(v_i, v_j) = 0$ for $i \neq j$ and Var $(v_i) =$ Var $(v_j) = 1$ for $i = 1, \ldots, b$ and $j = 1, \ldots, b$, i.e. the covariance matrix of the v's is just the identity matrix. The matrix K of the required transformation, i.e. $\mathbf{v} = K\boldsymbol{\omega}$, can be readily obtained in the following manner. Suppose that a set of elementary operations (cf. MACDUFFE,[19] pp. 31–33) $O_1, \ldots, O_Q$ in that order are performed on the rows of the covariance matrix of the ω's in order to change it to a triangular form in which all the diagonal elements are unity. If I_j denotes the matrix resulting from performing O_j on the identity matrix then $K = I \ldots I_2 I_1$. Denote the ith row, jth column element of K as K_{ij}. The weighted mean $\bar{\omega}$ can be computed with the relation

$$\bar{\omega} = \sum_{i=1}^{b} \left(\sum_{j=1}^{b} K_{ij} \right) \left(\sum_{j=1}^{b} K_{ij} \omega_j \right) \div \sum_{i=1}^{b} \left(\sum_{j=1}^{b} K_{ij} \right)^2 \qquad (65)$$

and this value used to calculate

$$\chi^2 = \sum_{i=1}^{b} (v_i - \bar{\omega} \sum_{j=1}^{b} K_{ij})^2 \qquad (66)$$

with $b - 1$ degrees of freedom. The value of

$$\chi^2 = \sum_{j=1}^{b} {v_i}^2 \qquad (67)$$

with b degrees of freedom should also be calculated. If equation (66) is non-significant for $b \geqslant 3$, one can infer that the autocorrelation effect is not important over the range of time intervals examined while if it is significant one must conclude that a^2 is appreciable in comparison to ${\sigma_m}^2/{\sigma_s}^2$. If (66) is non-significant but (67) is significant then one must conclude that since a^2 is not making an appreciable contribution to the value of ${\rho_z}^2 > 0$ it must be true that ${\sigma_m}^2/{\sigma_s}^2 > 0$. If this is so one must conclude that selection of high threshold animals by the first dose is playing a role in the mortality response to the second. If both (66) and (67) are non-significant one can conclude that thresholds separated by times of the order of magnitude of those to which the data pertains are statistically independent of one another.

For sufficiently large τ_j one would expect to find ${a_j}^2$ approaching zero and ${\rho_j}^2$ approaching ${\sigma_m}^2/({\sigma_m}^2 + {\sigma_s}^2)$ as a limit. Since ${\sigma}^2 = {\sigma_m}^2 + {\sigma_s}^2$ one should, in principle, be able to estimate ${\sigma_m}^2$ and ${\sigma_s}^2$ by the relations

$${\sigma_m}^2 = \sigma^2 \lim_{\tau_i \to \infty} {\rho_j}^2$$

$${\sigma_s}^2 = \sigma^2 (1 - \lim_{\tau_j \to \infty} {\rho_j}^2).$$

A possibility apt to foil this method of separate estimation of σ_s and σ_m in practice is that, for τ_j long enough to cause ρ_j to approach its limit, the assumption of a stationary time series for the thresholds and the assumption that $\alpha(t) = \alpha(t + \tau)$ might not be fulfilled.

There is a somewhat less obvious, but nonetheless serious, objection to the use of excessively long interdose intervals, full discussion of which will have to be deferred to another paper.[5] It was assumed that an animal whose

threshold is not exceeded at the time that r attains its first maximum will not have a threshold value less than r at any time prior to the time that r passes through its second maximum. It can be demonstrated (BEST, 1957) that, for sufficiently large values of τ_j, there will be a non-zero probability, increasing with increasing τ_j, of this occurring. This effect is a direct consequence of assuming a permanent component of the injury and the threshold to be a stochastic variable in time.

SUMMARY

Experiments to investigate the lethal effects of ionizing radiation are of three main types:

(1) Single dose, acute, using the proportion killed in a fixed time interval following the irradiation as an indicator of radiation damage.
(2) Duration of life, chronic, using the effect on the mean life span as an indicator of radiation damage.
(3) Fractionated dose experiments that may be regarded as intermediate between the first and second types.

The general problem of maximum likelihood estimation of the parameters of a formal prediction model in the case of a simple experimental design in which the dose is fractionated into two equal doses is discussed. A simple model is proposed in which the radiation is assumed to produce two additive components of injury, one of which is recoverable and the other permanent. The recoverable fraction of the injury is assumed to be repaired at a rate proportional to the remaining amount of recoverable injury present at any time. The formal relation between the injury and the radiation dose rate is thus identical with that previously suggested by BLAIR[6,7], SMITH[24] and SACHER.[23] A departure from the treatments of these authors is made in the assumption that the logs of the lethal thresholds of this injury are bivariate normally distributed with a correlation ρ_z that depends upon the time of separation of the two dose fractions. The formulae are derived for conducting the iterative computation to obtain the maximum likelihood estimates and the variance-covariance matrix of the estimates of the parameters of this model. An approximate method is derived for correcting for the variation in lag time between the infliction of fatal injury and death and for correcting for the normal death rate. A test of "goodness of fit" and a criterion for terminating the iteration are suggested. The relationship between ρ_z and the autocorrelation of the time series of the lethal threshold of an individual animal is discussed along with methods for ascertaining whether the autocorrelation or selection effects are more important in determining the relative probabilities of survival. Discussion of the relationship between proportion kill and shortened life span criteria of injury is deferred to another paper.[5]

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APPENDIX

The values of $\psi_2(h, a)$ and $\psi_3(h, a)$ are only tabulated for positive values of h and a. To obtain the values of these functions for negative values of h and a the following transformations are useful. The absolute magnitude of a shall be denoted here in this appendix as $\bar{a}$ and that of h as $\bar{h}$. Then

$$\text{for } a > 0,\ h > 0 \qquad \psi_2(h, a) = \psi_2(\bar{h}, \bar{a})$$
$$a < 0,\ h > 0 \qquad = -\psi_2(\bar{h}, \bar{a})$$
$$a > 0,\ h < 0 \qquad = -\psi_2(\bar{h}, \bar{a})$$
$$a < 0,\ h < 0 \qquad = \psi_2(\bar{h}, \bar{a})$$

The function $\psi_3(h, a)$ is even with respect to h and a, i.e.

$$\psi_3(h, -a) = \psi_3(h, a)$$
$$\psi_3(-h, a) = \psi_3(h, a)$$

The value of ψ_2 for $h = 0$ or $a = 0$ is zero.

Tabulated values of $\psi_2(h, a)$

a/h	0.1	0.2	0.3	0.4	0.5
0.1	0.001584	0.003120	0.004564	0.005875	0.007020
0.2	0.003167	0.006239	0.009124	0.01174	0.01422
0.3	0.004750	0.009355	0.01367	0.01759	0.02099
0.4	0.006333	0.01247	0.01821	0.02341	0.02790
0.5	0.007915	0.01557	0.02274	0.02919	0.03475
0.6	0.009496	0.01868	0.02724	0.03492	0.04151
0.7	0.01108	0.02177	0.03172	0.04061	0.04817
0.8	0.01265	0.02485	0.03617	0.04622	0.05472
0.9	0.01423	0.02793	0.04059	0.05177	0.06113
1.0	0.01581	0.03099	0.04497	0.05724	0.06741
1.1	0.01738	0.03405	0.04931	0.06262	0.07352
1.2	0.01896	0.03708	0.05361	0.06790	0.07948
1.3	0.02053	0.04011	0.05787	0.07309	0.08525
1.4	0.02210	0.04312	0.06207	0.07817	0.09084
1.5	0.02367	0.04611	0.06623	0.08313	0.09624
1.6	0.02523	0.04908	0.07032	0.08798	0.10144
1.7	0.02679	0.05204	0.07436	0.09271	0.10644
1.8	0.02835	0.05497	0.07834	0.09731	0.11123
1.9	0.02991	0.05788	0.08225	0.10178	0.11581
2.0	0.03146	0.06078	0.08610	0.10612	0.12018

Tabulated values of $\psi_2(h, a)$

a/h	0.6	0.7	0.8	0.9	1.0
0.1	0.007971	0.008713	0.009236	0.009541	0.009637
0.2	0.01591	0.01738	0.01841	0.01900	0.01918
0.3	0.02380	0.02597	0.02747	0.02832	0.02853
0.4	0.03160	0.03443	0.03636	0.03740	0.03761
0.5	0.03929	0.04273	0.04502	0.04620	0.04633
0.6	0.04684	0.05082	0.05341	0.05465	0.05462
0.7	0.05423	0.05868	0.06149	0.06270	0.06244
0.8	0.06142	0.06628	0.06921	0.07031	0.06972
0.9	0.06844	0.07358	0.07655	0.07744	0.07645
1.0	0.07522	0.08057	0.08347	0.08407	0.08260
1.1	0.08177	0.08723	0.08997	0.09018	0.08816
1.2	0.08805	0.09353	0.09602	0.09577	0.09314
1.3	0.09407	0.09948	0.10163	0.10085	0.09756
1.4	0.09982	0.10506	0.10679	0.10541	0.10144
1.5	0.10528	0.11027	0.11151	0.10949	0.10482
1.6	0.11046	0.11511	0.11580	0.11310	0.10773
1.7	0.11534	0.11959	0.11967	0.11628	0.11021
1.8	0.11994	0.12370	0.12314	0.11904	0.11229
1.9	0.12424	0.12748	0.12623	0.12143	0.11404
2.0	0.12827	0.13091	0.12897	0.12348	0.11548

Tabulated values of $\psi_2(h, a)$

a/h	1.1	1.2	1.3	1.4	1.5
0.1	0.009541	0.009274	0.008626	0.008335	0.007722
0.2	0.01897	0.01842	0.01758	0.01651	0.01527
0.3	0.02817	0.02730	0.02600	0.02437	0.02249
0.4	0.03704	0.03580	0.03401	0.03178	0.02924
0.5	0.04550	0.04384	0.04150	0.03863	0.03541
0.6	0.05345	0.05131	0.04838	0.04485	0.04092
0.7	0.06086	0.05817	0.05460	0.05038	0.04574
0.8	0.06766	0.06437	0.06012	0.05520	0.04985
0.9	0.07383	0.06989	0.06495	0.05932	0.05330
1.0	0.07937	0.07475	0.06910	0.06277	0.05611
1.1	0.08428	0.07895	0.07260	0.06561	0.05835
1.2	0.08857	0.08254	0.07551	0.06791	0.06011
1.3	0.09229	0.08556	0.07788	0.06972	0.06144
1.4	0.09547	0.08807	0.07979	0.07112	0.06244
1.5	0.09815	0.09012	0.08130	0.07219	0.06318
1.6	0.10038	0.09177	0.08247	0.07299	0.06370
1.7	0.10223	0.09308	0.08336	0.07357	0.06406
1.8	0.10373	0.09411	0.08403	0.07398	0.06431
1.9	0.10494	0.09490	0.08453	0.07410	0.06448
2.0	0.10590	0.09550	0.08488	0.07448	0.06458

Tabulated values of $\psi_2(h, a)$

a/h	1.6	1.7	1.8	1.9	2.0
0.1	0.007050	0.006348	0.005639	0.004944	0.004279
0.2	0.01392	0.01251	0.01110	0.009713	0.008392
0.3	0.02045	0.01834	0.01622	0.01415	0.01219
0.4	0.02650	0.02368	0.02086	0.01813	0.01556
0.5	0.03196	0.02843	0.02494	0.02158	0.01843
0.6	0.03677	0.03255	0.02842	0.02446	0.02078
0.7	0.04089	0.03602	0.03128	0.02679	0.02264
0.8	0.04434	0.03885	0.03356	0.02859	0.02404
0.9	0.04715	0.04110	0.03532	0.02995	0.02506
1.0	0.04938	0.04283	0.03664	0.03092	0.02577
1.1	0.05111	0.04413	0.03759	0.03161	0.02624
1.2	0.05242	0.04508	0.03826	0.03207	0.02655
1.3	0.05338	0.04575	0.03871	0.03236	0.02674
1.4	0.05407	0.04621	0.03901	0.03255	0.02686
1.5	0.05455	0.04652	0.03920	0.03266	0.02692
1.6	0.05488	0.04672	0.03932	0.03273	0.02696
1.7	0.05510	0.04684	0.03939	0.03277	0.02698
1.8	0.05524	0.04692	0.03943	0.03279	0.02699
1.9	0.05533	0.04697	0.03945	0.03280	0.02699
2.0	0.05538	0.04699	0.03946	0.03280	0.02699

Tabulated values of $\psi_3(h, a)$

a/h	0	0.1	0.2	0.3	0.4	0.5
0	0.1592	0.1584	0.1560	0.1521	0.1469	0.1405
0.1	0.1575	0.1568	0.1544	0.1506	0.1453	0.1389
0.2	0.1530	0.1522	0.1499	0.1460	0.1408	0.1344
0.3	0.1460	0.1452	0.1429	0.1390	0.1338	0.1274
0.4	0.1372	0.1364	0.1340	0.1302	0.1250	0.1187
0.5	0.1273	0.1265	0.1242	0.1204	0.1152	0.1089
0.6	0.1170	0.1162	0.1139	0.1101	0.1050	0.09873
0.7	0.1068	0.1060	0.1037	0.09989	0.09481	0.08867
0.8	0.09705	0.09625	0.09392	0.09014	0.08512	0,07906
0.9	0.08793	0.08714	0.08481	0.08105	0.07608	0.07012
1.0	0.07958	0.07879	0.07646	0.07273	0.06781	0.06197
1.1	0.07202	0.07122	0.06520	0.06520	0.06035	0.05463
1.2	0.06523	0.06444	0.05845	0.05845	0.05366	0.04808
1.3	0.05917	0.05838	0.05242	0.05242	0.04770	0.04227
1.4	0.05377	0.05298	0.04706	0.04706	0.04243	0.03714
1.5	0.04897	0.04818	0.04231	0.04231	0.03776	0.03262
1.6	0.04471	0.04392	0.03809	0.03809	0.03363	0.02865
1.7	0.04091	0.04013	0.03434	0.03434	0.02997	0.02516
1.8	0.03754	0.03675	0.03102	0.03102	0.02674	0.02208
1.9	0.03452	0.03374	0.02806	0.02806	0.02388	0.01939
2.0	0.03183	0.03104	0.02542	0.02542	0.02134	0.01704
2.1	0.02942	0.02863	0.02306	0.02306	0.01908	0.01496
2.2	0.02725	0.02647	0.02095	0.02095	0.01708	0.01314
2.3	0.02530	0.02452	0.01907	0.01907	0.01530	0.01153
2.4	0.02354	0.02276	0.01737	0.01737	0.01371	0.01011
2.5	0.02195	0.02117	0.01584	0.01584	0.01229	0.00887

Tabulated values of $\psi_3(h, a)$

a/h	0.6	0.7	0.8	0.9	1.0
0	0.1329	0.1246	0.1156	0.1062	0.09653
0.1	0.1314	0.1230	0.1141	0.1047	0.09510
0.2	0.1269	0.1186	0.1097	0.1004	0.09096
0.3	0.1200	0.1118	0.1030	0.09390	0.08467
0.4	0.1113	0.1033	0.09466	0.08577	0.07682
0.5	0.1017	0.09374	0.08535	0.07676	0.06816
0.6	0.09162	0.08387	0.07573	0.06743	0.05930
0.7	0.08169	0.07415	0.06630	0.05839	0.05069
0.8	0.07224	0.06494	0.05745	0.04992	0.04272
0.9	0.06348	0.05644	0.04929	0.04224	0.03559
1.0	0.05552	0.04875	0.04198	0.03539	0.02928
1.1	0.04838	0.04189	0.03552	0.02942	0.02384
1.2	0.04204	0.03589	0.02986	0.02428	0.01926
1.3	0.03645	0.03061	0.02502	0.01991	0.01542
1.4	0.03157	0.02605	0.02086	0.01622	0.01225
1.5	0.02727	0.02208	0.01731	0.01314	0.00964
1.6	0.02356	0.01868	0.01432	0.01058	0.00754
1.7	0.02032	0.01577	0.01178	0.00847	0.00585
1.8	0.01749	0.01329	0.00967	0.00674	0.00451
1.9	0.01506	0.01116	0.00789	0.00534	0.00345
2.0	0.01294	0.00935	0.00642	0.00421	0.00261
2.1	0.01110	0.00782	0.00521	0.00329	0.00197
2.2	0.00952	0.00651	0.00421	0.00256	0.00147
2.3	0.00815	0.00541	0.00338	0.00198	0.00109
2.4	0.00697	0.00449	0.00271	0.00152	0.00080
2.5	0.00595	0.00371	0.00216	0.00117	0.00058

Tabulated values of $\psi_3(h, a)$

a/h	1.1	1.2	1.3	1.4	1.5
0	0.08691	0.07747	0.06837	0.05973	0.05167
0.1	0.08557	0.07615	0.06716	0.05856	0.05062
0.2	0.08155	0.07235	0.06353	0.05521	0.04747
0.3	0.07555	0.06660	0.05815	0.05015	0.04284
0.4	0.06799	0.05955	0.05149	0.04401	0.03724
0.5	0.05976	0.05174	0.04431	0.03742	0.03120
0.6	0.05138	0.04398	0.03709	0.03085	0.02535
0.7	0.04335	0.03653	0.03032	0.02480	0.01998
0.8	0.03597	0.02978	0.02426	0.01945	0.01534
0.9	0.02941	0.02390	0.01905	0.01491	0.01148
1.0	0.02372	0.01886	0.01467	0.01121	0.00839
1.1	0.01892	0.01467	0.01112	0.00826	0.00599
1.2	0.01491	0.01127	0.00829	0.00597	0.00419
1.3	0.01162	0.00853	0.00610	0.00423	0.00286
1.4	0.00896	0.00638	0.00440	0.00295	0.00192
1.5	0.00686	0.00472	0.00314	0.00203	0.00127
1.6	0.00518	0.00345	0.00221	0.00136	0.00081
1.7	0.00388	0.00248	0.00153	0.00090	0.00051
1.8	0.00289	0.00177	0.00104	0.00059	0.00032
1.9	0.00212	0.00125	0.00070	0.00038	0.00019
2.0	0.00154	0.00087	0.00047	0.00024	0.00011
2.1	0.00111	0.00060	0.00030	0.00015	0.00007
2.2	0.00080	0.00041	0.00020	0.00009	0.00004
2.3	0.00056	0.00027	0.00012	0.00005	0.00002
2.4	0.00039	0.00018	0.00008	0.00003	0.00001
2.5	0.00027	0.00012	0.00005	0.00002	0.00001

Tabulated values of $\psi_3(h, a)$

a/h	1.6	1.7	1.8	1.9	2.0
0	0.04425	0.03752	0.03150	0.02618	0.02154
0.1	0.04326	0.03665	0.03068	0.02548	0.02090
0.2	0.04040	0.03022	0.02837	0.02340	0.01910
0.3	0.03621	0.03022	0.02499	0.02040	0.01651
0.4	0.03110	0.02567	0.02094	0.01692	0.01348
0.5	0.02570	0.02090	0.01682	0.01334	0.01045
0.6	0.02052	0.01642	0.01293	0.01004	0.00771
0.7	0.01586	0.01241	0.00956	0.00726	0.00543
0.8	0.01189	0.00908	0.00681	0.00503	0.00365
0.9	0.00866	0.00643	0.00468	0.00335	0.00235
1.0	0.00615	0.00442	0.00311	0.00215	0.00145
1.1	0.00425	0.00296	0.00201	0.00134	0.00087
1.2	0.00287	0.00192	0.00125	0.00080	0.00050
1.3	0.00189	0.00121	0.00076	0.00046	0.00027
1.4	0.00122	0.00075	0.00044	0.00026	0.00014
1.5	0.00076	0.00045	0.00025	0.00014	0.00007
1.6	0.00047	0.00026	0.00014	0.00007	0.00004
1.7	0.00028	0.00015	0.00007	0.00004	0.00002
1.8	0.00016	0.00008	0.00004	0.00002	0.00001
1.9	0.00009	0.00004	0.00002	0.00001	0.00000
2.0	0.00005	0.00002	0.00001	0.00000	0.00000
2.1	0.00003	0.00001	0.00000	0.00000	0.00000
2.2	0.00002	0.00001	0.00000	0.00000	0.00000
2.3	0.00001	0.00000	0.00000	0.00000	0.00000
2.4	0.00000	0.00000	0.00000	0.00000	0.00000
2.5	0.00000	0.00000	0.00000	0.00000	0.00000

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THEORETICAL RELATIONSHIP BETWEEN PERCENT KILL AND SHORTENED LIFE SPAN MEASURES OF RADIATION INJURY

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Abstract—In a companion paper[1] a mathematical model and maximum likelihood estimation procedure were presented for the analysis of data regarding the percentage kill from fractionated doses of ionizing radiation. It was postulated that the logarithm of an animal's lethal threshold at two different times is bivariate normally distributed with a correlation whose magnitude will, in general, be less than unity. In the present paper this manner of interpreting kill data is explored in respect to certain of its theoretical implications concerning life-span effects. Although conceived merely as a two dose analogy to the rationale used to interpret single dose kill experiments this postulate introduces the possibility that an animal whose lethal threshold is not exceeded by the maximum injury inflicted during the acute phases following radiation exposure may, at some later time, die by a stochastic transition of its threshold from a value greater to one less than the permanent injury residual from that exposure. This implies that even among those animals which have survived the acute phase, there will occur an increased probability of death per unit time with a subsequent decrease in their mean life-span. This property is a likely consequence of any mathematical model in which a permanent injury component and random variation in time of the lethal threshold of an individual are features of the theory.

Two different types of measures have been extensively used to assess the injury produced in mammals by whole body dosage with ionizing radiation. These two types of measures are: (1) The proportion of animals dying within some fixed time interval after exposure to a given dose of radiation. (2) The effect of radiation on the mean life span of the irradiated animals.

The various formal theories proposed to account for the lethal effects observed with different dose schedules of whole body irradiation define injury in terms of one or the other of these measures, but do not relate the two.

In another paper[1] a formal theory of radiation injury was derived for predicting the percentage mortality from certain types of whole body fractionated dose experiments. Death from radiation injury was postulated to occur if and only if an injury function r exceeded a threshold value x of an animal. The functional dependence of r upon the rate of dosage $\gamma(t)$ and the time t was assumed to be similar to those previously proposed by BLAIR,[2,3] SMITH[10] and SACHER,[9] i.e. that

$$r(t)=A\int_0^t \gamma(t')\,dt' + (1-A)e^{-Bt}\int_0^t e^{Bt'}\gamma(t')\,dt'$$

where A is a dimensionless number between zero and one. It is assumed that $\gamma = 0$ for $t < 0$. An assumption was made concerning the distribution of thresholds, however, that is a departure from the discussions of these authors. It was postulated that the logarithms of two successive measurements of x, i.e. x_1 at t_1 and x_2 at t_2, made on each animal, will be bivariate normally distributed with the same standard deviation σ and mean μ for each of the two variates and correlation ρ between them. The correlation ρ was postulated to be dependent upon the absolute magnitude of the difference

$t_2 - t_1$ but, aside from this, not dependent upon the values of t_2 or t_1. The correlation will approach unity as $t_2 - t_1$ approaches zero. An experimental situation was considered in which a pair of doses of radiation is given to each animal in such a manner that the time interval during which a dose is administered is small and the two doses separated by a waiting period of length τ. Three time intervals were defined such that death in the first interval could be primarily ascribed to the result of the first dose and death in the second interval primarily ascribed to the additive effects of the two doses. An animal surviving the first two intervals would be classified into the third interval. These three intervals are exhaustive. A method for the calculation of the maximum likelihood estimates of the parameters of the radiation injury model from such kill data was derived.

This formulation of the relation between lethal effects and whole body irradiation, however, permits an interpretation of measurements of type (2) in terms of the same set of parameters and model used to interpret type (1). In the previous paper,[1] this point was merely alluded to and not actually demonstrated. It will be the main purpose of the present paper to derive expressions permitting the interpretation of type (2) data in terms of this same model with the hope that this will lead to clarification of the relationship between these two types of data.

If the actuarial function, relating the probability of death of the members of a population to time and dosage schedule, is known then, clearly, such particular characteristics as mean life-span are strictly determined. It will therefore be the main objective of this paper to obtain such an actuarial function in terms of the model and parameters previously mentioned.

It will be convenient to make the following definitions with respect to notation:

$$R = \frac{\log r - \mu}{\sigma}\,;\quad X = \frac{\log x - \mu}{\sigma}\,;$$

denote the value of R at t_1 as R_1 and at t_2 as R_2; $\Delta t = t_2 - t_1$; $\Delta R = R_2 - R_1$;

$\Pr\{(E_1) \cdot (E_2)\}$ = joint probability of occurrence of the events E_1 and E_2;

$\Pr\{(E_2) \mid (E_1)\}$ = probability of occurrence of E_2 given E_1;

$\Pr\{E_1\}$ = probability of E_1;

$j(X_1, X_2; \rho)$ = bivariate normal density function with means at zero, standard deviations of unity, correlation ρ and stochastic variables X_1 and X_2;

$$G(h) = \frac{1}{\sqrt{(2\pi)}} \int_{-\infty}^{h} \exp\left(-\frac{y^2}{2}\right) dy$$

$$T(h, a) = \frac{1}{2\pi} \int_0^a \frac{\exp\left\{-\frac{h^2}{2}(1 + y^2)\right\}}{1 + y^2}\, dy.$$

Consider the probability that $X_1 > R_1$ at t_1 and $X_2 < R_2$ at t_2 where $t_2 > t_1$. If Δt is sufficiently small then multiple transitions (back and forth) can be ignored so that $\Pr\{(X_1 > R_1) \cdot (X_2 < R_2)\}$ will be the probability of single transition in Δt. The probability that an individual, known to have $X_1 > R_1$, will undergo transition to a state in which $X_2 < R_2$ in the interval Δt will therefore be

$$\Pr\{(X_2 < R_2) \mid (X_1 > R_1)\} = \frac{\Pr\{(X_1 > R_1) \cdot (X_2 < R_2)\}}{\Pr\{X_1 > R_1\}}. \quad (1)$$

Clearly, according to the postulates of the model, this will be the probability that an individual, alive at t, will be dead of radiation injury by $t + \Delta t$ if the lag between death and infliction of fatal injury is neglected. If n is the number of animals alive at t then

$$-\frac{1}{n}\frac{dn}{dt} = \phi(t) \quad (2)$$

where

$$\phi(t) = \lim_{\Delta t \to 0} \left(\frac{\Pr\{(X_1 > R) \cdot (X_2 < R + \Delta R)\}}{\Delta t \cdot \Pr\{X_1 > R\}}\right) \quad (3)$$

and deaths from causes other than radiation injury are ignored. The task then becomes essentially that of deriving the expression for $\phi(t)$.

$$\Pr\{(X_1 > R_1) \cdot (X_2 < R_2)\} = \int_{R_1}^{\infty} \int_{-\infty}^{R_2} f(X_1, X_2;\ \rho)\, dX_1\, dX_2 = \bar{M}(R_1, R_2; \rho). \quad (4)$$

For sufficiently small Δt and finite values for the rate of radiation dosage, ΔR will be small so that the probability integral can be written in the form

$$\bar{M}(R_1, R_2; \rho) = \bar{M}(R, R; \rho) + (\Delta R) \cdot \lim_{\Delta R \to 0} \frac{\partial \bar{M}(R_1, R_2; \rho)}{\partial R_2}. \quad (5)$$

From the results of OWEN[7] one can write that

$$\bar{M}(R_1, R_2; \rho) = \tfrac{1}{2}G(R_2) - \tfrac{1}{2}G(R_1) + T\left(R_1, \frac{R_2 - \rho R_1}{R_1\sqrt{(1-\rho^2)}}\right) + T\left(R_2, \frac{R_1 - \rho R_2}{R_2\sqrt{(1-\rho^2)}}\right) \quad \text{if} \quad R_1 R_2 > 0 \quad (6)$$

or $R_1 R_2 = 0$ and R_1 or $R_2 > 0$, or R_1 and $R_2 = 0$. Thus

$$\bar{M}(R, R; \rho) = 2T\left\{R, \sqrt{\left(\frac{1-\rho}{1+\rho}\right)}\right\} \quad (7)$$

and

$$\lim_{\Delta R \to 0} \frac{\partial \bar{M}(R_1, R_2; \rho)}{\partial R_2} = \frac{\exp\left(-\frac{R^2}{2}\right)}{\sqrt{(2\pi)}}\, G\left\{R\sqrt{\left(\frac{1-\rho}{1+\rho}\right)}\right\}. \quad (8)$$

Substituting expressions (7) and (8) into expression (5) one obtains

$$\bar{M}(R, R + \Delta R; \rho) = 2T\left\{R, \sqrt{\left(\frac{1-\rho}{1+\rho}\right)}\right\} + \frac{\exp\left(-\frac{R^2}{2}\right)}{\sqrt{(2\pi)}}\, G\left\{R\sqrt{\left(\frac{1-\rho}{1+\rho}\right)}\right\} \Delta R. \quad (9)$$

Expression (9) was obtained on the assumption that (6) was valid. If ΔR is sufficiently small and $R_1 > 0$ or $R_1 < 0$, then $R_1 R_2 > 0$. If $R_1 = 0$ then, in the limit as $\Delta R \to 0$, R_1 and R_2 will both be zero. Thus, although for more than infinitesimal differences between R_1 and R_2 expression (6) may not be appropriate, in the limit it will always be.

Similarly, the function $T\left\{R, \sqrt{\left(\frac{1-\rho}{1+\rho}\right)}\right\}$ can be expanded in terms of powers of $\sqrt{\left(\frac{1-\rho}{1+\rho}\right)}$, noting that $\sqrt{\left(\frac{1-\rho}{1+\rho}\right)} \to 0$ as $\Delta t \to 0$. Define

$$C = \sqrt{\left(\frac{1-\rho}{1+\rho}\right)}.$$

Then $T(R, C) = T(R, 0) + \dfrac{\exp\left(-\frac{R^2}{2}\right)}{2\pi} C \quad (10)$

for small C. Substituting (10) into (9) and noting that $T(R, 0) = 0$ one obtains

$$\bar{M}(R, R + \Delta R; \rho) = \frac{\exp\left(-\frac{R^2}{2}\right)}{\pi} C + \frac{\exp\left(-\frac{R^2}{2}\right)}{\sqrt{(2\pi)}}\, G(RC) \cdot \Delta R. \quad (11)$$

Since $P\{X_1 > R_1\} = 1 - G(R_1)$ one can write

$$\phi(t) = \lim_{\Delta t \to 0} \left\{\frac{\bar{M}(R, R + \Delta R; \rho)}{(1 - G(R)) \cdot \Delta t}\right\}. \quad (12)$$

Noting that $\lim_{\Delta t \to 0} \dfrac{C}{\Delta t} = \lim_{\Delta t \to 0} \dfrac{dC}{d(\Delta t)}$; $\lim_{\Delta t \to 0} \dfrac{\Delta R}{\Delta t} = \dfrac{dR}{dt}$; $\lim_{\Delta t \to 0} C = 0$ and $G(0) = \tfrac{1}{2}$

one obtains that

$$\phi(t) = \frac{1}{(1 - G(R))} \left[\frac{\exp\left(-\frac{R^2}{2}\right)}{\pi} \lim_{\Delta t \to 0} \frac{dC}{d(\Delta t)} + \frac{\exp\left(-\frac{R^2}{2}\right)}{2\sqrt{(2\pi)}} \frac{dR}{dt}\right]_+ \quad (13)$$

where

$$[y]_+ = y \quad \text{if} \quad y > 0 \qquad = 0 \quad \text{if} \quad y \leqslant 0 \quad (14)$$

It can be demonstrated (see Appendix) that in order for

$$\lim_{\Delta t \to 0} \frac{dC}{d(\Delta t)}$$

to exist it is necessary and sufficient to require that the functional dependence of ρ upon Δt be such that

$$\lim_{\Delta t \to 0} \frac{d\rho}{d(\Delta t)} = 0 \text{ and } \lim_{\Delta t \to 0} \left(\frac{d^2\rho}{d(\Delta t)^2} \right) < 0. \quad (15)$$

With the stipulation of conditions (15) on ρ one finds that

$$\lim_{\Delta t \to 0} \frac{dC}{d(\Delta t)} = \left(\frac{\xi}{2} \right)^{\frac{1}{2}} \quad (16)$$

and $\rho \doteq 1 - \xi \cdot (\Delta t)^2$ for sufficiently small Δt, where ξ is a positive number.

Thus

$$\phi(t) = \frac{\exp\left(-\frac{R^2}{2} \right)}{2\sqrt{(2\pi)}} \frac{\left[2\left(\frac{\xi}{\pi} \right)^{\frac{1}{2}} + \frac{dR}{dt} \right]_+}{(1 - G(R))}. \quad (17)$$

The implications of relation (17) will be discussed later on after the correction for the normal death rate has been obtained.

Let $\alpha(t)$ be the death rate due to natural causes. According to BRUES and SACHER,[4] the GOMPERTZ[6] function gives an adequate description of the natural mortality, thus in this case

$$\alpha(t) = \alpha_0 / \exp(mt) \quad (18)$$

where α_0 and m are constants. It will be assumed that death from aging is an independent process from that produced by radiation injury. Then

$$-\frac{1}{n}\frac{dn}{dt} = \alpha_0 \exp\{m(t + t_0)\} + \frac{\exp\left(-\frac{R^2}{2} \right)}{2\sqrt{(2\pi)}} \times \frac{[2\sqrt{(\xi/\pi)} + dR/dt]_+}{(1 - G(R))} \quad (19)$$

where t_0 is the age of the population at the beginning of the irradiation, t is the time after onset of the irradiation and the lag between the fatal event and actual occurrence of death is ignored.

One more item deserves mention. Suppose that a high rate of dosage of radiation is administered over a short interval of time, e.g. 10 r/min for 10 min. The amount of recovery occurring in this short interval of time will be negligible so that for practical purposes one can say that 100 r were administered instantaneously. Stated in this way, however, the derivative dR/dt is infinite and $R(t)$ has a discontinuity. If the animal has previously been uninjured by radiation then the discontinuity in R will be infinite. If the animal has been previously injured then the discontinuity in R will be finite. It is advantageous to be able to incorporate such discontinuities into the formalism since for practical purposes it will make very little difference whether 5 r/min are delivered for 20 min or 10 r/min are delivered for 10 min.

The following notational device will be helpful in the discussion of such discontinuities. Suppose a discontinuity in R occurs at some time t_1 then the value of R immediately preceding t_1 shall be denoted as R_{b1} and the value immediately following t_1 as R_{a1}. A similar notation can be used to denote the number whose lethal thresholds have not been exceeded, i.e. let n_{b1} and n_{a1}, respectively, denote the numbers with thresholds greater than their injury immediately before and immediately after t_1. Thus for such a discontinuity at t_1, the number killed at the discontinuity would be

$$n_{b1}\left(\frac{G(R_{a1}) - G(R_{b1})}{1 - G(R_{b1})} \right) \quad (20)$$

and the number surviving

$$n_{b1}\left(\frac{1 - G(R_{a1})}{1 - G(R_{b1})} \right). \quad (21)$$

To examine the effect of such discontinuities it will be instructive to consider a simple conceptual experiment. Suppose that a dose schedule is arranged (for details of such a schedule see the Appendix) so

$$\left.\begin{aligned} &R = -\infty, \text{ i.e. } r = 0 \text{ for } t < 0 \\ &R = R_0 = \text{const} \quad \text{for } 0 < t < t_1 \\ &R = R_1 = \text{const} \quad \text{for } t > t_1 \end{aligned}\right\}. \quad (22)$$

The injury R will thus be constant aside from discontinuous steps at $t = 0$ and $t = t_1$. For the moment let us neglect the deaths occurring from natural causes in the interval $0 < t < t_1$. Let n_{b0} be the number of animals used for the experiment which will also be the number alive just prior to $t = 0$. The number of animals

killed at the first discontinuity will be $n_{b0}G(R_0)$, while the number killed at the second will be

$$n_{b1} - n_{a1} = n_{b1}\left(\frac{G(R_1) - G(R_0)}{1 - G(R_0)}\right). \quad (23)$$

During the interval 0 to t_1 some animals will die by undergoing transitions of the sort described by equation (17). Thus, the number of such transition deaths occurring during the interval $0 < t < t_1$ will be

$$n_{a0} - n_{b1} = n_{b0}$$

$$\left(1 - \exp\left\{-\frac{e^{-R_0^2/2}}{\pi(1 - G(R_0))}\sqrt{\frac{\xi}{2}}\,t_1\right\}\right). \quad (24)$$

The total number of deaths produced by the two discontinuities and the transitions during $0 < t < t_1$ will then be

$$n_{a1} - n_{b0} = n_{b0}\left[1 - (1 - G(R_1)) \times \exp\left\{-\frac{e^{-R_0^2/2}}{\pi(1 - G(R_0))}\sqrt{\frac{\xi}{2}}\,t_1\right\}\right]. \quad (25)$$

In the event that $t_1 = 0$ the total number of deaths becomes $n_{b0}G(R_1)$, or just that number expected from a single dose producing a sudden injury of R_1. It is interesting to note also from expression (24) that the probability of death becomes greater the longer the interval t_1 even though natural death has been neglected and the injury remains constant throughout the interval 0 to t_1. This effect originates entirely from the temporally stochastic character ascribed to the thresholds and the transition deaths arising thereby.

A case of interest is that in which a single dose of radiation is administered within an interval of time of negligible duration. Suppose that D r are so administered to animals of age t_0 that have not been previously exposed to radiation. R and dR/dt will have the values

$$R = R_0 = \frac{1}{\sigma}(\ln D - \mu) \qquad \text{at } t = 0$$

$$R = R_0 + \frac{1}{\sigma}\ln(A + (1-A)e^{-BT}) \quad \text{at } t > 0$$

$$\frac{dR}{dt} = -\frac{B(1 - A)e^{-Bt}}{\sigma \cdot (A + (1 - A)e^{-Bt})} \quad \text{at } t > 0.$$

At $t = 0$ there will be a discontinuous step in R resulting in the death of $n_0G(R_0)$ of the animals. Let n be the number of animals still surviving at a time t after the radiation was administered. This can be obtained from equation (19) with appropriate attention to the effect of the discontinuity at $t = 0$. Thus

$$\ln\frac{n}{n_0} = \ln(1 - G(R_0)) - \frac{\alpha_0}{m}(e^{m(t+t_0)} - e^{mt_0}) - \frac{1}{2\sqrt{(2\pi)}}\int_0^t \frac{e^{-R^2/2}}{(1 - G(R))}\left[2\sqrt{\left(\frac{\xi}{\pi}\right)} - \frac{B(1 - A)e^{-Bt'}}{(A + (1 - A)e^{-Bt'})}\right]_A dt'. \quad (26)$$

In the treatment of the paired dose experiment[1] two assumptions were made that deserve discussion at this point. It was assumed that the second term on the right in equation (26) could be represented by a quantity linear in t. This will bean adequate approximation only for sufficiently short intervals of time since for small t this term reduces to the form $\alpha_0 \exp(mt_0)t$. The third term on the right involves the contribution due to stochastic transition and was also neglected in the treatment presented in the other paper.[1] The magnitude of the contribution from this term likewise depends on the length of the time interval under consideration. The fact that $dR/dt < 0$ during the intervals of primary interest for the other paper will lessen the effect of this term. In the case that long intervals are under consideration, however, this term will become appreciable and must be considered. The effect of this term leads to an interesting consequence that might be inferred from the earlier discussion of the present paper. Suppose a group of animals are given such a single dose D and one then considers the n^* survivors still remaining at a time t^* such that $t^* \gg 1/B$. At this time the recovery process will be complete and the acute aspects of the dose effects over with. The number n for $t > t^*$ will be given by the expression

$$\ln\frac{n}{n^*} = -\frac{\alpha_0}{m}(e^{-mt} - e^{-mt^*}) - \frac{1}{\pi}\sqrt{\left(\frac{\xi}{2}\right)}\int_{t^*}^t \frac{e^{-(R^*)^2/2}dt'}{(1 - G(R^*))} \quad (27)$$

where $R^* = R_0 + \frac{1}{\sigma} \ln A$

Since R^* is independent of the time, equation (27) can be readily integrated to obtain

$$\ln \frac{n}{n^*} = \frac{\alpha_0}{m} (e^{-mt} - e^{-mt^*}) - \frac{\sqrt{(\xi)}\, e^{-(R^*)^2/2}\, (t - t^*)}{\pi \sqrt{(2)(1 - G(R^*))}}. \quad (28)$$

It is clear from equation (28) that the mortality rate among these "survivors" will still be higher than normal and result in a decreased life expectancy for them. This effect on life expectancy, it should be noted, was not produced by an *ad hoc* equating to the residual injury but arises as a logical consequence of the treatment of the acute kill, i.e. type 1, data.

It is, perhaps, interesting to note that this effect on life span arises in a way that resembles certain recent views of SACHER[9] concerning the lethal event. SACHER has suggested that the physiological state of an animal varies with time in a manner that resembles a random walk in the presence of restoring forces. The various homeostatic devices, acting with "negative feedback" to restore the state of the organism to an optimal condition, are considered by him to be reflected in these restoring forces. If the state of the animal exceeds some lethal bound then the animal dies.

The equations used in the present treatment are not the same as those whose properties were investigated by SACHER[9] but the qualitative features and philosophical viewpoint arrived at are nevertheless similar. In the present treatment the results arose from a rather modest formulation intended for use in the description of kill data from two-step fractionated dose experiments. The model suggested in this and the companion paper[1] is about the simplest and most obvious extension of the rationale (e.g. see FINNEY,[5] pp. 8–36) used in the analysis of type 1 data that can be made. However, as we have seen, this leads directly to a consideration of effects of long duration and, coincident with the views of SACHER,[9] the possibility of death by stochastic transition.

As more data from fractionated dose experiments become available, it may well be the case that the model presented in this and the companion paper will not be adequate. For example, it may turn out that some other unimodal bivariate probability distribution must be used for the thresholds or that the differential equations giving the temporal decrement of the injury are too simple. Nevertheless, if the modified mathematical model contains a term corresponding to the permanent component of the injury and the bivariate threshold distribution is such that there exists a non-zero probability that the lethal threshold of a surviving animal will take on (during an interval of time of the order of magnitude of the average life-span) a value less than or equal to the value of the permanent component, an effect on the life-span of the "survivors", similar to that discussed in the present treatment, will be implied.

One final aspect of the present theory should be indicated. By insisting that the variance σ^2 of the threshold distribution be the same at two successive times one is, in effect, saying that if the stochastic time series describing the lethal threshold is a continuous random walk then it is a random walk in the presence of restoring forces. If the successive values of the threshold in time were generated by an unconstrained random walk then the variance of the threshold distribution would be a monotonically increasing function of time. This would mean that the threshold variance for older animal populations would always be greater than that of younger animal populations. In so far as one can ascertain there is no evidence that this is so.

APPENDIX I

To achieve a situation in which the injury is effectively a step function of the time, one can administer radiation according to the following dose schedule. A dose of $\gamma_0/B(1 - A)$ r is delivered within a short interval of time of length Δt. At the same time that this dosage is going on, and thereafter, an additional amount of radiation is administered at the rate $\gamma_0 e^{-ABt}$. The total rate of administration of radiation during the initial short interval

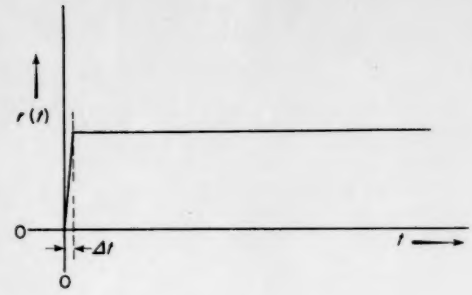
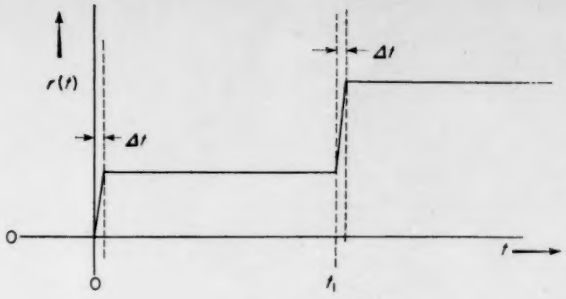
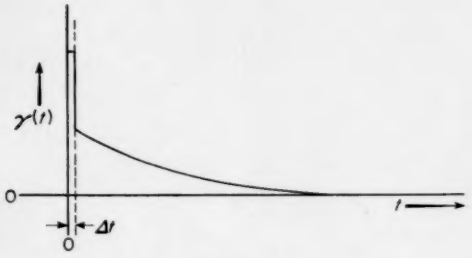


FIG. 1.





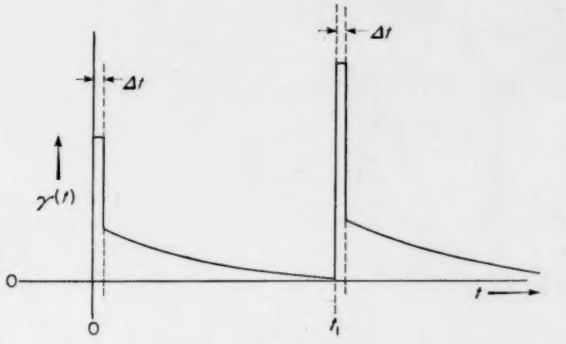


FIG. 2.

from $t = 0$ to $t = \Delta t$ will then be

$$\gamma(t) = \frac{\gamma_0}{B(1 - A) \cdot \Delta t} + \gamma_0 e^{-ABt} \quad \text{where } B \cdot \Delta t \ll 1$$

and γ_0 is an arbitrary dosage constant. For all $t > \Delta t$ deliver radiation at the rate

$$\gamma(t) = \gamma_0 e^{-ABt}.$$

It will be noted that, since $(1 - A)$ is less than 1 and $B \cdot \Delta t \ll 1$, $\gamma_0/B(1 - A) \cdot \Delta t$ will be much greater than γ_0.

The rate, γ, of administration of the radiation dosage and the injury, r, as functions of time will thus appear somewhat as shown in Fig. 1.

In a similar manner a radiation schedule can be arranged so that the resultant injury is effectively a two-step function with one step at $t = 0$ and another at t_1.

This result will be obtained if

for $t < 0$

$$\gamma(t) = 0$$

for $0 \leqslant t \leqslant \Delta t$ where $\Delta t \ll 1/B$

$$\gamma(t) = \frac{\gamma_0}{B(1 - A)\Delta t} + \gamma_0 e^{-ABt}$$

for $\Delta t < t < t_1$

$$\gamma(t) = \gamma_0 e^{-ABt}$$

for $t_1 \leqslant t \leqslant t_1 + \Delta t$

$$\gamma(t) = \frac{\gamma_1}{B(1 - A)\Delta t} + \gamma_1 e^{-AB(t - t_1)} + \gamma_0 e^{-ABt}$$

for all $t > t_1 + \Delta t$

$$\gamma(t) = \gamma_1 e^{-AB(t - t_1)} + \gamma_0 e^{-ABt}.$$

With this schedule the dose rate $\gamma(t)$ and the injury $r(t)$ as functions of time will appear somewhat as shown in Fig. 2. In this case γ_0 and γ_1 are arbitrary constants whose values are determined by the size of the steps required at $t = 0$ and $t = t_1$.

APPENDIX II

Consider the quantity $C = \sqrt{\left(\frac{1 - \rho}{1 + \rho}\right)}$ when ρ refers to the correlation of two threshold

values of the same animal at two times t_1 and t_2. Suppose that $\Delta t = t_2 - t_1$ is small. As $\Delta t \to 0$, $\rho \to 1$, since the two thresholds must become identical in the limit if the threshold of a particular animal at a given time is to be unique. For some sufficiently small neighborhood of Δt about the value $\Delta t = 0$ one can consider ρ to be of the form

$$\rho = 1 + k_s \cdot (\Delta t)^s$$

where s is a positive number and k_s is the first non-zero coefficient in the power series expansion of ρ. If Δt is sufficiently small then terms involving powers of Δt greater than s can be ignored. One can write

$$C = \left(\frac{-k_s \cdot (\Delta t)^s}{2 + k_s(\Delta t)^s}\right)^{\frac{1}{s}}$$

for sufficiently small Δt. Differentiating with respect to Δt and letting Δt become small, one obtains

$$\frac{dc}{d(\Delta t)} = \frac{s}{2^{3/2}} (k_s)^{\frac{1}{s}} (\Delta t)^{\frac{s}{2} - 1}.$$

If $\frac{s}{2} < 1$ then $\lim_{\Delta t \to 0} \frac{dc}{d(\Delta t)} = \infty$ while

if $\frac{s}{2} > 1$ then $\lim_{\Delta t \to 0} \frac{dc}{d(\Delta t)} = 0$. Thus, if

the limit is to exist and be non-trivial, it is clear that $s/2$ must be equal to 1, or $s = 2$. Also, if this limit is to be real then k_s must be negative otherwise $(-k_s)^{\frac{1}{s}}$ will be imaginary. Hence the limiting form of ρ must be

$$\rho = 1 - \xi \cdot (\Delta t)^2$$

where $\xi = -k_s$ is a positive number. For this to be the limiting form of the powers expansion in Δt it is necessary and sufficient that the first derivative of ρ with respect to Δt vanish and that the second derivative be negative for $\Delta t = 0$.

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STRAY RADIATION DOSIMETRY AROUND PULSED ACCELERATORS*

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Abstract—The dosimetry of stray radiation fields around pulsed accelerators is complicated by the fact that the radiation comes in pulses and by the mixed nature of the radiation usually produced by these accelerators. Suitable instrumentation for pulsed radiation consists of special uses of standard ratemeters and counters and integrating type ion chamber instruments. Fixed monitors and personnel monitoring schemes must be adapted to this special use.

If a mixture of radiation is to be measured with ionization chambers, the physical dose can be obtained if the chamber is tissue equivalent, but an effective or weighted average value of r.b.e. must be determined to obtain the biological dose. An experimental procedure using a cloud chamber has been developed to measure the distribution of l.e.t. Stereoscopic photographs of the tracks in the cloud chamber are taken. The length of every track is measured and the optical density classified. By calibration with tracks of known particles, the energy deposited in the sensitive volume from different l.e.t. classes can be calculated. An effective r.b.e. is obtained from values of r.b.e. as a function of l.e.t. as tabulated in *NBS Handbook* 59.

INTRODUCTION

When monitoring the health physics hazards in the stray radiation fields around a pulsed accelerator, two complications are encountered which are not generally found in other dosimetry studies. The first is the fact that the radiation is produced in pulses. This makes the instrumentation and the data interpretation difficult. The second is that the radiation which such accelerators usually produce, is a complicated mixture. This results in difficult dose determinations.

The simplest type of pulsed accelerator is one in which a steady beam is interrupted to produce pulsed radiation. Whenever possible, this type is best surveyed under steady beam conditions with the application of appropriate correction factors obtained from the constants of the pulsing operation. This procedure is not possible, however, if the pulsing is a necessary feature for the operation of the accelerator.

The energy of the primary particles from a pulsed accelerator is often high, with the result that the stray radiation is a mixture of particles and γ-rays produced by nuclear reactions of the primary particles with surrounding matter. The operating constants and maximum energy of a few pulsed accelerators are shown in Table 1.[1] From the ranges in pulse repetition rate and pulse duration shown, it is evident that each accelerator presents a special problem.

BASIC APPROACH

Two approaches to the evaluation of the health hazard of mixed radiation are possible.[2] The first is the measurement of each component of the radiation independently and the summation of the biological doses from each. The second is the measurement of the total dose in rads by the use of a tissue equivalent ionization chamber and the application of an effective or weighted average relative biological effectiveness (r.b.e.) to provide the biological dose.

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Table 1

	Rep. rate (pulses/sec)	Pulse Dur. (μsec)	Energy (MeV)	Type of particle
NRL	180	2	23	e
Stanford	60	0·6	70	e
Minn.	15	200	68	p
Purdue	4	100	300	e
Bevatron	$\frac{1}{4}$	10,000	6400	p
Cosmotron	$\frac{1}{5}$	10,000	3000	p

The effective r.b.e., which must be determined by other measurements, will depend on the fractional amount of dosage due to each type of radiation and the value of r.b.e. appropriate for each. It will, therefore, be a constant as long as the mixture of radiation is constant.

The appropriate choice between these two methods depends on the accelerator. In some cases it can be established that for routine monitoring the measurement of one component is sufficient to establish the hazard. For instance, the neutron hazard from some accelerators so greatly outweighs the associated γ-hazard that only neutron surveys are necessary. However, the very high energy proton accelerators produce stray radiations in which there is no predominant type. Short-lived particles such as mesons, V-particles, etc., for which no specific detectors exist, are produced copiously by such machines as the Cosmotron and Bevatron. To separate the various types of radiation obtained from these accelerators is a complicated experiment which is unsuited from a practical standpoint for daily survey work. In these cases, ionization chamber measurements can be used for surveying.

The surveying of the Cosmotron offers an example of the use of both of these two systems. Routine surveys are based on ionization chamber measurements, but in order to assign a quantitatively determined value of effective r.b.e., work is being done to determine the quality of the stray radiation. The choice of this r.b.e. may always be deliberately made too high; for instance, that of the most hazardous type of radiation present may be used. Additional costs in shielding and manpower will result from such an over-conservative choice. The proper choice of r.b.e. can be made only if the flux and energy distribution of the different types of radiation are determined, or if the distribution of the linear energy transfer (l.e.t.) of the ionizing particles is known. Either of these involved determinations leads to a practical value of r.b.e. Since the neutron hazard is the most serious, O'Brien has done some work at the Cosmotron to determine the neutron flux and energy distribution using various threshold detectors.[3,4] Thermal neutron fluxes have been measured with activation foils and Oleson determined a crude neutron energy distribution from the activation of these foils at various depths in paraffin.[5] In this paper, work will be reported on the use of a cloud chamber to measure the l.e.t. distribution.

The effective r.b.e. in areas accessible to personnel at the Cosmotron is estimated to be no greater than 5. In view of some uncertainties involved in this estimate a safety factor of 2 has been used which increases the operating r.b.e. to 10.

SURVEY INSTRUMENTS

Any instruments used around pulsed accelerators must first satisfy the requirements for use around any accelerator. These include the elimination of magnetic and r.f. influences on the instrument. Often r.f. interference can be shielded with screening or aluminum foil, and magnetic fields shielded with soft iron. Magnetic fields usually affect photomultipliers very seriously, but if the magnetic

fields are not too strong the commercially available shields for these tubes usually suffice.

The instantaneous dose rate in the stray radiation fields around pulsed accelerators is usually not known, but can be very high. If the duty cycle and the average dose rate are known, then the average dose rate over the time of the pulse can be calculated. However, this dose rate is usually not constant but a peak of radiation generally occurs at some time in the pulse. Factors which can change the instantaneous dose rate in relation to the average dose rate over the time of the pulse are location, accelerator operation and shielding. At the Cosmotron the pulse of radiation has a duration of about 1 sec. During the acceleration period, radiation is produced all around the machine and when the beam intercepts the target, the instantaneous dose rate increases by at least a factor of 2. This factor depends on location and is higher than 2 in the vicinity of the target. Usually it is not necessary to measure the instantaneous dose rates, but detectors must be operated in such a manner as to be reliable at the highest instantaneous dose rates encountered. Experiments to determine the saturation voltage of ionization chambers and the dead time of counters must be made. The accelerator itself can serve as a source to determine suitable ion chamber voltages and complete saturation curves can be taken at the accelerator even though the instantaneous and average dose rates are not known. The dead time of a counter will, when compared to the pulse duration and the absolute counting rate in the pulsed radiation, determine its suitability.

Standard survey instruments are sometimes adequate for pulsed radiation. However, the interpretation of the readings and the use of such instruments must be made carefully. An example of such use was reported by BRODSKY using NRL's Betatron.[6,7] This accelerator has a repetition rate of 180 pulses/sec and a pulse duration of 2 μsec. Because of the short pulse length, and the relatively long dead time and low efficiency of the BF_3 counter used, the counter cannot register more than 1 count/pulse. The maximum counting rate is therefore, 180 c/s. In high radiation fields which produce about 1 count/pulse or greater this detector is useless, but the efficiency is such that at health hazard levels the counting rate is much less than 1 count/pulse and its counting rate is, in fact, shown to be proportional to the dose rate over a limited range. The calibration of this instrument made with steady radiation is good for this particular pulsed radiation if the limit of this proportional region of operation is not exceeded.

COSMOTRON SURVEY INSTRUMENTS

A special use has been made at the BNL Cosmotron of commercial ionization chamber ratemeters. Because of the time parameters of this accelerator these survey instruments can be used ballistically. Each pulse of radiation produces a ballistic deflection of the meter, the maximum of which is proportional to the charge from the chamber. This proportionality exists because the pulse-length is short compared to the time constant of the instrument. The time between pulses (5 sec) is long enough to allow the meter to return to zero. Two commercial instruments which have the desired characteristics for this use are the Beckman MX–2 and the Nuclear Research & Development CS–40. Care must be taken to assure saturation of the chamber. The 67 V collecting potential commonly found in ratemeters of this type must be increased. It has been found that even if relatively high levels of radiation are to be measured, 300 V will saturate the chambers. Calibration of the instruments is performed at the Cosmotron by comparison with integrating instruments; the original calibration is obviously ignored. In general, each instrument, even of the same type, will read differently when used in this manner. In the steady radiation case, the feedback circuit is devised to make the meter reading almost independent of the gain of the electrometer tube and the constants of the feedback circuit, and to reduce the time constant of the instrument. When used ballistically, however, the meter reads the change in feedback voltage, which is dependent on the gain of the electrometer and on the feedback circuit constants. The amplifier essentially behaves as if little or no feedback were present, and the meter reads the amplifier signal directly. However, even a small feedback voltage will have a large effect on the meter reading. Consequently small differences in the feedback circuits of similar instruments can result in serious differences in the sensitivity to pulsed radiation. Each instrument must thus be individually calibrated. These difficulties could be avoided by properly designing instruments for this particular use.

The standard survey instrument at the Cosmotron is an integrating meter. It consists of an ionization chamber and electrometer circuit[8] with the high value grid resistor removed and the grid left floating on a low value capacitance. This circuit integrates the charge from the chamber over any measured time interval. Since the repetition rate of the Cosmotron is low (one pulse every 5 sec) the integration is done over a given number of pulses. A shorting switch is provided to discharge the input capacitance when necessary. Insulation

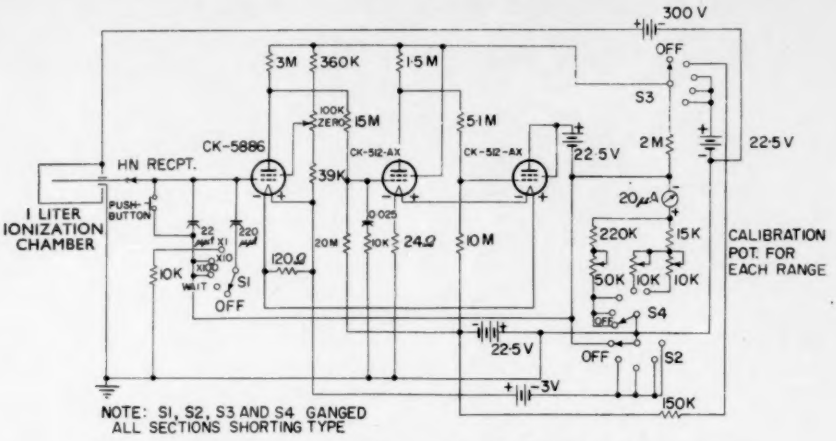

FIG. 1. Schematic diagram of integrating ion chamber survey meter.

requirements are more critical than with similar ratemeter electrometer circuits because a small leakage current results in a drift whereas in the case of the ratemeter a leakage produces a small constant deflection which can be zeroed out. Calibration of the instruments is done with standard Co^{60} sources and the most sensitive scale is adjusted so that if one pulse produces full scale deflection, the average radiation level is 5 mrads/hr (or 50 mrems/hr) with a pulsing rate of 720 pulses/hr. Full scale deflection is equivalent to about 0.008 mrad. In order to attain this sensitivity, the input capacity must be small, that is from 10 to 25 pF. All of the insulation except that of the tube base and added capacitance is Teflon.

The collecting potential used for these chambers must be such that the chamber is saturated in the presence of the high instantaneous dose rates. This voltage has been experimentally determined by plotting saturation curves using the Cosmotron as a source. Calculations of the collection efficiency based on arguments presented in the literature[9] confirm the experimental results that the chambers are well saturated and that the collection efficiency is essentially 100 per cent when the collecting potential is 300 V. Various sized chambers have been used to obtain instruments of different sensitivities.

A new type integrating instrument has been developed by the BNL Instrumentation Division. The basic instrument has been described by HIGINBOTHAM[10] but the circuit has been modified to integrate the input charge and a guard ring has been added to the chamber. The circuit is shown in Fig. 1. Three ranges of sensitivity are obtained from switching two input capacities and the meter circuit. All switching of the input circuit is done at low impedance and consequently leakage between the grid circuit and ground is independent of the leakage of the switch. This is accomplished by connecting one end of the larger capacitor either to ground or the feedback loop. When it is connected to the feedback loop the net integrating capacitance is that of the two capacitors in parallel; when it is connected to ground, its contribution is reduced by the gain–feedback product. The open loop gain of the circuit is about 1000 so that even with the large capacitor grounded the gain–feedback product is 100. The instrument case includes a discharging switch controlled by a camera cable release mounted on the handle. Pulsed radiation dose measurements made with this type of instrument are independent of the repetition rate and the pulse duration of the accelerator.

Ideally, these chambers, which are made with bakelite walls, should have tissue equivalent walls and filling gas. Comparative measurements have been made with tissue equivalent ion chambers[11] and the bakelite chambers and correction factors obtained for different locations at the Cosmotron. The dose measurements used in practice are therefore based on these tissue equivalent ionization chamber measurements.

Cosmotron fixed monitors

In addition to portable survey instruments, fixed monitors are used. The Cosmotron control room is monitored with an integrating ion chamber, vibrating reed electrometer and chart recorder. The sensitivity is adjusted so that full scale deflection

of the recorder is 0.1 mrad, or 1.0 mrem. The charging condenser and chamber are discharged with a solenoid when the recorder reaches full scale. Each traverse of the recorder is registered on a mechanical counter, the reading of which gives directly the total mrem exposure. This type of instrument is also used with the tissue equivalent chamber to compare the tissue equivalent and bakelite chambers. The ratios of doses measured with tissue equivalent chambers to those measured with bakelite chambers vary between 1 and 1.7, depending on location.

Personnel monitoring

Film badges are convenient integrators of dose and, for that reason, are very important around pulsed accelerators. Because of the large experimental areas associated with high energy machines, large areas are often irradiated with low level dose rates, with the result that doses to individuals working in these areas are difficult to predict or calculate from surveys. If a mixture of radiation exists, the interpretation of the densities behind various filters bears careful consideration. At high energies it is possible that reactions occur in the filter material, and no high energy calibrated sources exist with which to standardize the film densities. Calculation of the expected absorption from filters is difficult because cross-section data for some filter materials is incomplete. If neutrons are present, nuclear track film can be used as the personnel dosimeter, but the film should be calibrated for use in a mixed field of radiation. The number of tracks observed for a given biological dose will depend on the energy of the neutrons and the amount of other types of radiation present which will not be recorded by the nuclear track film.

At the Cosmotron, NTA track counting is the basis for all personnel dose records. The calibration is performed at the Cosmotron using the tissue equivalent chamber as a reference. The number of tracks constituting a maximum weekly exposure is not the same as that observed from Po–Be neutrons, because the dose from radiations other than neutrons or protons is not recorded by the NTA film, but is recorded by the chamber. In general if neutrons are present, nuclear track film is a good indicator of the biological dose received from accelerators, but the absolute number of tracks corresponding to a maximum permissible exposure will vary with factors such as energy, location and amount and type of shielding.

Determination of effective r.b.e.

In order to successfully use ionization chamber measurements in mixed radiation fields a value of effective r.b.e. must be chosen. An experiment has been devised therefore to measure this quantity. This experiment uses a cloud chamber and by examination of the tracks produced in the chamber, an approximate value of the effective r.b.e. can be obtained. A relationship between r.b.e. and l.e.t. is given in *NBS Handbook* 59[12] for use in health hazard evaluation studies. Fig. 2 shows this relationship for tissue equivalent gas. The general scheme of the experiment is to determine the l.e.t. distribution from data obtained from the tracks and then by weighting this distribution with the appropriate r.b.e.'s, obtain an effective r.b.e.

The diffusion cloud chamber used was furnished

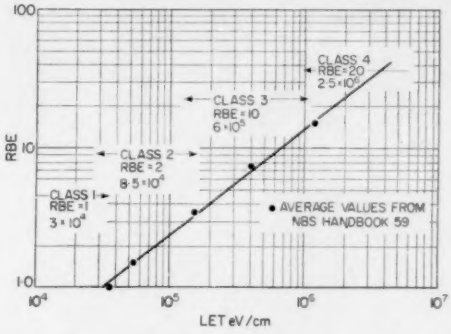

FIG. 2. Dependence of r.b.e. on l.e.t. (*NBS Handbook* 59) for tissue equivalent gas.

by the BNL Cloud Chamber Group, and is equipped with a camera to take stereoscopic pictures of the tracks. It has been exposed in the stray radiation areas around the Cosmotron and photographs taken of every pulse by triggering the arc lights by a signal from the Cosmotron. The cloud chamber is filled with one atmosphere of tissue equivalent gas (64.4 per cent methane, 32.5 per cent CO_2, 3.1 per cent N_2; percentages by partial pressure). With a special projection system, the length of any track photographed is measured directly, regardless of its angle with respect to the sensitive volume. The optical density of a track is related to the l.e.t. of the particle and this relationship can be observed by exposing the cloud chamber to known radiations. A secondary from a γ-ray produces a track with the lowest specific ionization, while an α-particle produces a very dense track. The optical densities of these tracks of known particles, with known energy, are compared with the tracks from exposures at the Cosmotron. The l.e.t. of the known tracks is obtained from Fig. 3[13] and, hence, the l.e.t. of the unknown tracks can be inferred.

In practice, four classes of densities are chosen and each track is visually classified into one of these classes. The l.e.t. ranges of the classes are designated in Fig. 2. All the tracks are measured and the total length of track in each class is determined. The average l.e.t. and the total length of track for each class are used to determine the energy loss associated with each class of track. By weighting this energy loss with the average r.b.e. for that class, and adding the contribution from all classes, the effective r.b.e. is obtained.

Preliminary data are shown in Table 2. An effective r.b.e. of approximately 8 resulted from the analysis of one hundred photographs. This effective r.b.e. is computed by first calculating the energy associated with each class. This energy is the product of the l.e.t. for that class and the measured length. The fraction of the total energy occurring in each class is then calculated. The sum of the products of these fractions and the appropriate values of r.b.e. is the weighted average or effective r.b.e. The data tabulated in Table 2 were taken in the experimental area adjacent to the external pencil beam and are considered only as an indication that the experiment is feasible. Since the cloud chamber is steel and large masses of metal surround the sensitive volume, the results are hardly applicable to tissue.

Steady radiation and background tracks can occur at any time, and can therefore be photographed as "old" tracks which appear denser than newly formed ones. An expansion type cloud chamber would eliminate this difficulty because the tracks would all be the same age. From the experience

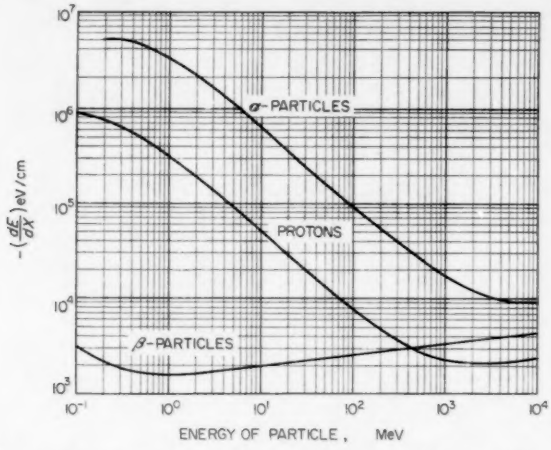

FIG. 3. Linear energy transfer for various particles in tissue equivalent gas as a function of particle energy (15°C, 760 mm).

Table 2

Class	Avg. l.e.t. (eV/cm)	Measured length (cm)	Avg. r.b.e.	Energy (eV)	Fraction of total energy	Fraction ×r.b.e.
1	3.4×10^4	1450	1	5.0×10^7	0.3	0.3
2	8.5×10^4	300	2	2.5×10^7	0.15	0.3
3	6.0×10^5	97	10	5.8×10^7	0.35	3.5
4	2.5×10^6	13.5	20	3.4×10^7	0.2	4.0
				Eff. r.b.e. =		8.1

obtained from this preliminary experiment, plans have been made to construct an expansion type cloud chamber with plastic walls. Possibly tissue equivalent plastic will be used. It is hoped that this chamber will be ready for operation when the major repairs now being made to the Cosmotron are completed.

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PHYSIOLOGY AND TOXICOLOGY OF PLUTONIUM-239 AND ITS INDUSTRIAL MEDICAL CONTROL*

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Abstract—When taken into the systemic circulation, Pu^{239} deposits predominantly in the skeleton, where it may produce bone disease (including cancer) many years later. Its absorption rate from the gastrointestinal tract is only about 0.003 per cent. A small amount may be absorbed through the intact skin and through contaminated cuts and puncture wounds. Absorption from the lung may be from 1 to 10 per cent of the inhaled dose, depending on particle size, solubility, chemical form, etc. Inhalation of contaminated air is potentially the most important mode of exposure, and its control is largely responsible for the rigorous closed-systems and other industrial hygiene and engineering practices employed in plutonium processing. Once in the body, Pu^{239} is excreted extremely slowly (about 200 years being required to eliminate one-half the body burden). An individual who has reached the maximum permissible body burden technically should be removed from further plutonium contact for the rest of his life. The maximum permissible body burden of Pu^{239} (0.04 μc) is established by comparison with Ra^{226} and is that amount which has the same improbability of producing harm to any person at any time during his natural life as does 0.1 μc of fixed Ra^{226}.

Control of the industrial hazards of Pu^{239} processing is based on the premise that exposure of personnel should be as nearly zero as possible. This is not because less than the maximum permissible body burden is apt to do harm, but because it is sound industrial medical and economic practice. If presently recommended practices are maintained, there is little reason to feel that the health of a person working with Pu^{239} will be subject to any greater absolute risk than if he were engaged in any other chemical or industrial occupation.

INTRODUCTION

During the past 15 years, processing of Pu^{239} has grown from novel microtechniques applied by a few individuals into a routine industrial procedure involving many people and kilogram quantities of material. Concurrent with the growth of the industry has been a progressive realization that the most effective control of the industrial medical problems of plutonium processing is through the adoption of rigorous and elaborate industrial engineering measures such that exposure of operating personnel is as nearly zero as possible. This emphasis on minimal exposure conditions, unless viewed in context with the toxicological and physiological properties of plutonium, may cause unwarranted apprehension on the part of those engaged in plutonium work. This statement is not meant to belittle the absolute toxicity of plutonium in any manner. Animal experiments suggest that Pu^{239} deposited in bone is potentially more dangerous than Ra^{226}, and quite small amounts of radium have produced disabling and fatal bone disease in man.[1–4]

The physiological and toxicological properties of Pu^{239} are summarized in this report to provide better understanding of its potential as an industrial hazard and to explain further the necessity for rigorous industrial hygiene and engineering control over all plutonium processing.

RADIOACTIVE PROPERTIES OF PLUTONIUM-239

Plutonium^{239} decays by α-emission, emitting α-particles with an average energy of 5.15 MeV and having a range of about 40 μ in water and soft tissue. It emits several weak X-rays with

* Work done under the auspices of the U.S. Atomic Energy Commission.

energies of from 10 to 22 keV and a few 380 keV γ-rays. Material produced at high power levels emits fast neutrons ($\sim$1 MeV) from spontaneous fission of Pu^{240} and a negligible amount of 60 keV γ-radiation from Am^{240}. Heavily neutron-irradiated plutonium will, of course, give off β- and γ-radiation in relation to the quantity and age of the fission products present. The radiological half-life of Pu^{239} is 24,400 years. One microgram (1 μg) of this material is equivalent to 0.064 μc, or approximately 1.4×10^5 α-disintegrations per min.

PHYSIOLOGICAL PROPERTIES OF PLUTONIUM-239

Absorption

Gastrointestinal tract. Experiments on mature rats[5–9] and pigs[10,11] indicate that only from 0.01 to 0.003 per cent of an orally administered dose of Pu^{239} is absorbed into the blood stream. The extremely low gastrointestinal absorption rate appears to be essentially independent of the valence state of the plutonium and the amounts ingested[7,8] but somewhat dependent on the acidity of the plutonium solution.[8,12] That which is not absorbed is apparently hydrolyzed, adsorbed to the food residues, and passed out in the feces within 24 to 36 hr.

Intact skin. Absorption of Pu^{239} through the unbroken skin has been studied in man and in experimental. animals. The percentage of applied plutonium absorbed through the skin of rats was shown to be independent of the area of skin exposed and the plutonium concentration but dependent upon the acidity of the plutonium solution applied.[13–15] Over a 5 day period, about 0.3 per cent of the plutonium applied in 0.1 N nitric acid was absorbed. When the nitric acid was increased to 10 N, the absorption was approximately 2 per cent. The skin of the rat, however, is quite different from human skin, especially from that of the palm of the hand (which is the area most likely to be contaminated). An experiment on man in which 10 μg of plutonium as $Pu(NO_3)_4$ in 0.4 N nitric acid was applied to the palm and allowed to remain for 8 hr gave an absorption rate of approximately 0.0002 per cent per hr.[16]

Wounds. Plutonium salts may be introduced into the body through abrasions, cuts and punctures in the skin. Microgram amounts of plutonium oxide placed on freshly abraded areas of the skin of rabbits failed to show any absorption into the body, but the oxide was incorporated into the scab and was lost when the scab became detached.[17] Pieces of plutonium metal (ranging in size from 0.67 to 1.8 mg) implanted subcutaneously in rabbits and rats were rapidly oxidized, but most of the oxide remained localized at the site of implantation.[18] Absorption into the body ranged from 0.09 to 1.2 per cent of the implanted dose over the entire life span of the animal (260 to 1048 days). Absorption of soluble plutonium salts through skin lacerations in rats has been reported as being greater than through the intact skin.[19] SCOTT and co-workers[20] showed that intramuscular injection of Pu^{3+}, Pu^{4+}, and PuO_2^{2+} resulted in absorption of 23, 4 and 30 per cent, respectively, from the site of injection after a period of 4 days, indicating a slow rate of plutonium translocation to other organs and tissues of the body. These experiments, as well as observations of actual accidents occurring in processing operations, show that introduction of plutonium and its compounds into wounds in the skin and subcutaneous tissues (especially in the case of cuts and puncture wounds) can result in significant but slow absorption into the systemic circulation.

Lungs. The problem of lung absorption, retention and elimination of inhaled materials is almost hopelessly complex, since the various factors are dependent on particle size of the material inhaled[21], solubility,[22] particle density,[23] rate of respiration of the individual,[24] etc. Although it is not possible at present to determine quantitatively what happens to inhaled plutonium under all specific conditions of exposure, it is possible on the basis of animal experiments to make some broad generalizations.[25]

If 100 radioactive particles of optimum size for lung retention are inhaled, about 25 are immediately exhaled without depositing in the lungs. Of the 75 particles that remain in the lungs 50 are deposited in the bronchial tree and removed in a few hours to a few days by ciliary action and swallowed. Of the remaining 25 particles which were deposited in the alveolar

sacs, about 10 (10 per cent of the originally inhaled dose) are rather rapidly absorbed into the circulating blood and deposited predominantly in the skeleton. The remaining 15 particles may be phagocytized, deposited in the lymph nodes, or eliminated up the bronchial tree and swallowed, the time of removal being of the order of from 150 to 200 days. Experimental data on animals, at least in so far as plutonium oxide is concerned,[26] suggest that 10 per cent absorption may be a conservative upper limit. Absorption may actually be more nearly 1 per cent. Nevertheless, the data do suggest that inhalation is the principal potential route of entry of plutonium into the body. Experiments on animals show also that some of the inhaled material may accumulate in the pulmonary lymph nodes from which the elimination rate is slow, resulting several days later in a higher concentration in the nodes than in the lungs proper.[26,27]

Body deposition

Regardless of method of administration, plutonium is rapidly cleared from the blood stream and fixed in the tissues. Table 1 shows the rate of disappearance of Pu^{239} from the blood of man following intravenous administration of a small dose of Pu^{4+}-citrate. Approximately 65 per cent of the plutonium was removed from the blood in 4 hr, and about 85 per cent was removed in only 1 day. Plutonium in the blood is believed to be combined largely with the globulin fraction of the serum proteins, and regardless of the valence of the material administered it is believed to be all in the form of Pu^{4+} within a few hours.

Although the relative distribution of plutonium in the various tissues and organs is somewhat dependent on chemical form and method of administration, liver and skeleton are the major sites of deposition. When administered as a strong complex (e.g. citrate or versenate) or as PuO_2^{2+}, from 60 to 80 per cent of the dose is deposited in the skeleton and from 10 to 20 per cent in the liver. When injected as uncomplexed Pu^{3+} and Pu^{4+}, liver deposition is approximately 25 and 40 per cent and skeletal deposition approximately 45 and 30 per cent, respectively.[5] The greater liver uptake following intravenous injection of uncomplexed Pu^{3+} and Pu^{4+} may be explained by complex colloid formation at the pH of the blood, resulting in greater reticuloendothelial fixation.

Table 1. Plutonium-239 content of total blood volume as a function of time after administration*

Time after administration	Pu^{239} in blood (%)†
4 hours	36
1 day	16
2 days	10
3 days	8.6
6 days	3.4
10 days	1.2
15 days	0.7
22 days	0.4

* Total blood volume taken as 7.7 per cent of body weight.

† Percentage of administered dose.

Distribution of Pu^{239} in man about 5 months after injection of Pu^{239} complexed with citrate is shown in Table 2. About 90 per cent of the injected material was deposited in only two organs (67 per cent in the skeleton and 23 per cent in the liver). Slower entry of plutonium into the blood stream (e.g. via absorption from the gastrointestinal tract and the lungs) as opposed to intravenous injection may give greater deposition in the bone and less in the liver.[5] A few months after systemic exposure to plutonium, from 80 to 90 per cent of the total body burden may be found in the skeleton, where it is believed to be adsorbed on the surfaces of newly formed and rapidly metabolizing bone mineral.[28]

Autoradiographs of bone sections (Fig. 1) show the material deposited in a fine line paralleling the endosteal and periosteal surfaces with highly localized concentrations in regions of trabecular bone.[29] The deposition patterns of plutonium and radium, although grossly similar, are somewhat different microscopically. Because of its similarity to calcium, radium is chemically incorporated into bone mineral, giving a more diffuse deposition pattern with time than does plutonium. This basic difference

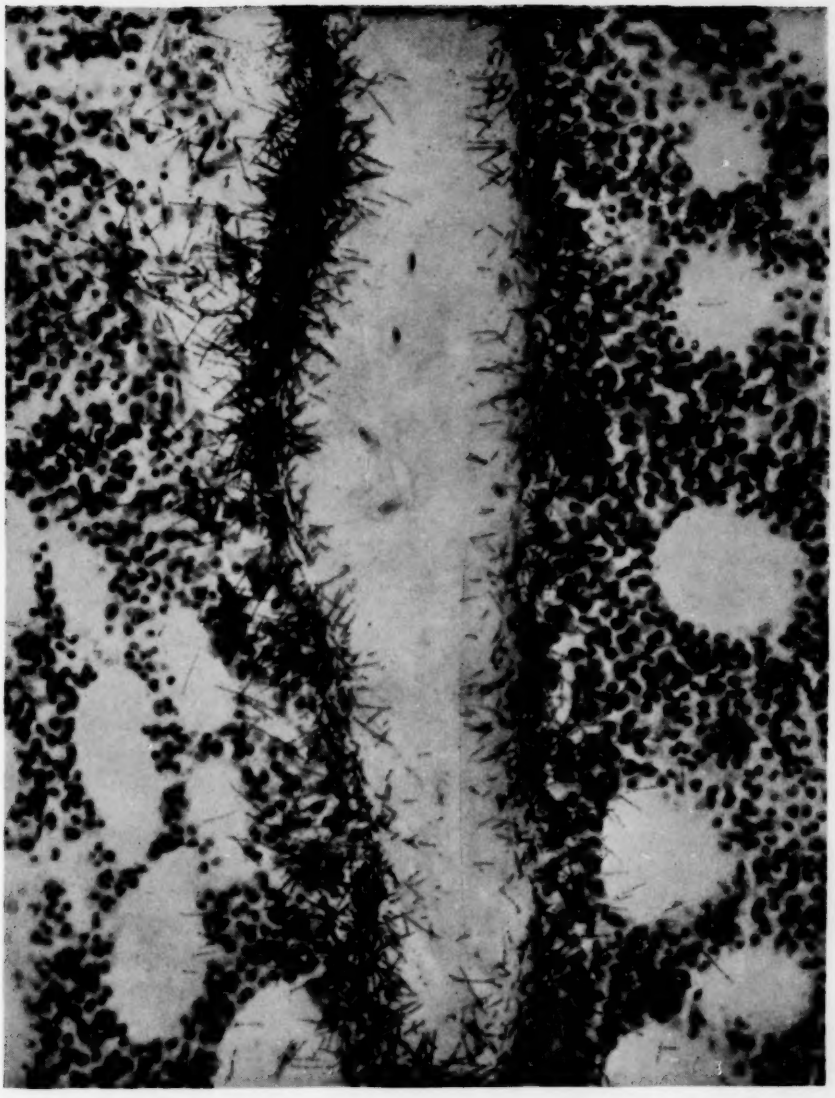

FIG. 1. Autoradiograph showing plutonium deposition on the surface of a trabecula of the proximal humoral head in a dog sacrificed 24 hr after injection.
(*Courtesy of W. S. S. Jee, Radiobiology Laboratory, University of Utah.*)

Table 2. Distribution of Pu^{239} *in man following injection of* Pu^{4+} *complexed with citrate*

Tissue or organ	Weight of organ or tissue (g)	Plutonium per organ (%)*
Skeleton (including marrow)	10,000	66
Liver	1700	23
Spleen	300	0.4
Kidneys	700	0.4
Lungs	1000	1.0
Lymphoid tissue	700	0.5
Heart	300	0.1
Gastrointestinal tract	2000	0.5
Muscle and skin	36,100	3.9
Blood	5400	0.2
Balance	11,800	1.0
Excreted (urine and feces)	—	5.0
Total	70,000	102

* Expressed as percentage of injected dose.

in deposition patterns of radium and plutonium is believed to be the principal reason for the greater relative radiotoxicity of the latter.

Excretion and retention

Excretion of Pu^{239} has been studied in rats,[5] dogs,[30] pigs[31] and man.[25] Although species variations occur, the rate of excretion by all species is slow. The plutonium elimination rate of the dog is comparable to that of man, and unlike man and the dog, rats eliminate fifteen times as much plutonium in the feces as in the urine. Fig. 2 shows the urinary and urinary plus fecal excretion of plutonium by man (over a period of approximately 5 years) as a function of time after administration. Empirically, the urinary excretion curve (curve II) fits the following power function:

$$Y_u = 0.20 \times t^{-0.74}, \qquad t > 1 \qquad (1)$$

in which Y_u is the percentage of the administered dose excreted per day, and t is the number of days between exposure and collection of the sample. A recent resurvey of the early Los Alamos plutonium exposure cases suggests that this expression holds reasonably well over a period of at least 12 years. The fecal excretion curve (not shown in Fig. 2) is fitted by the expression:

$$Y_f = 0.63t^{-1.09}, \qquad t > 1 \qquad (2)$$

in which Y_f is the percentage of the dose excreted per day in the feces. The total urinary plus fecal excretion rate (Y_{u+f}) shown by curve I in Fig. 2 is represented by the sum of the two expressions. The corresponding expression for total urinary plus fecal excretion of Ra^{226} by man was given by Norris *et al.*[32] as:

$$Y_{u+f} = 28t^{-1.52}, \qquad t > 1. \qquad (3)$$

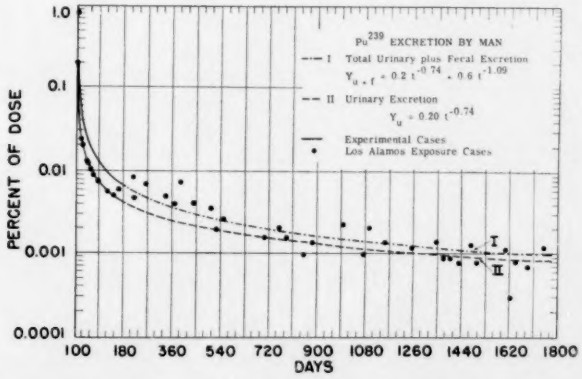

Fig. 2. Urinary and urinary plus fecal excretion of plutonium by man over a period of approximately 5 years.

A number of factors regarding the excretion and retention of plutonium by man are apparent from the above expressions. First, the plutonium excretion rate is not a simple exponential but becomes slower as the period of fixation becomes longer. That is to say, the elimination half-time increases with time. Second, the fecal to urinary excretion ratio is not constant. During the first 30 days, the ratio is slightly greater than 1, after which it drops to less than 1, becoming progressively smaller with time. Third, the excretion rate of plutonium is extremely low; less than 1 per cent is excreted during the first 24 hr after administration, compared to from 30 to 40 per cent for Ra^{226}. The rate of Pu^{239} excretion continues to decrease with time until only about 0.01 per cent is being excreted per day at about 100 days after exposure. At approximately 5 years, the rate has dropped to about 0.001 per cent. Table 3 shows the relative accumulated excretion of plutonium and radium at various times after exposure. These data were obtained by integrating the respective excretion expressions between the limits of 1 and t days after exposure. These data show that an individual may be expected to retain about 80 per cent of his original plutonium body burden 50 years after exposure, while he would be expected to retain only about 0.3 per cent of his original body burden of radium. Solution of the integral expression for 50 per cent excretion shows that approximately 200 years would be required for man to eliminate half his plutonium body burden.

Table 3. Accumulated urinary plus fecal excretion of Ra^{226} *and* Pu^{239} *by man*

Time after administration	Accumulated excretion (% administered dose)	
	Ra^{226}	Pu^{239}
1 day	46	0.5
10 days	84	2.6
50 days	93	4.0
100 days	95	4.7
1 year	97.5	6.3
3 years	98.6	7.8
5 years	98.9	8.7
10 years	99.2	10.0
20 years	99.5	12.2
50 years	99.7	17.6

RADIOTOXICOLOGY AND INDUSTRIAL MEDICAL CONTROL OF PLUTONIUM-239

The maximum permissible body burden of natural uranium, based on chemical toxicity to the kidney, is about 40 mg. Plutonium is chemically very similar to uranium and may be expected to have about the same chemical toxicity. Because of its greater specific activity (about 2×10^5 that of uranium), it may be considered entirely as a potential radiological hazard.

External radiation from plutonium-239. Plutonium-239 presents no potential external α-radiation hazard when deposited on the skin, since the α-particle range in tissue is about 40 μ and the cornified (dead) epithelium of the skin surface is from 70 to 150 μ thick. Plutonium-239 does, however, emit weak X-rays with an average energy of about 17 keV and some neutrons ($\sim$1 MeV energy), depending on the Pu^{240} content. The X-ray emission is much too soft to produce a general external hazard; however, the radiation dose rate to the skin of the hands, when in contact with unclad Pu^{239} metal, is about 1 rad/hr. Through heavy neoprene drybox gloves, the dose rate is about 400 mrads/hr, giving a maximum permissible weekly contact time of about 4 hr. Under usual conditions, the external radiation hazard from neutron emission is considered negligible. Plutonium that has been irradiated with neutrons will, of course, contain fission products which will present a β–γ radiation hazard that will be dependent on the quantity and age of the fission products present.

Plutonium-239 *contamination of the skin.* As mentioned above, there is no potential external alpha radiation hazard associated with Pu^{239} deposited on the intact skin surface. The possibility of a small but finite absorption rate of plutonium through the skin and from contaminated wounds and the possibility of transfer of activity from the hands to food, cigarettes, etc., make control of hand contamination a

good industrial hygiene practice. Wearing gloves while working with plutonium and routine hand monitoring twice daily are simple and effective control measures. Effective methods of skin decontamination are available. The standard Hanford procedure[33] of swabbing with liquid soap followed by a water rinse, and alternate $KMnO_4$ applications (4% solution) and $NaHSO_3$ (5% solution), is over 98 per cent effective. Versene (10% solution) is also very effective and less traumatizing to the skin.

Plutonium-239 *embedded locally in the tissues.* Plutonium introduced into the skin and subcutaneous tissues through lacerations and puncture wounds may remain at the site of the wound for a very long time. α-Radiation of the surrounding tissue, in this case, may produce local damage. Less than 1 μg of plutonium (0.064μc) as $Pu(NO_3)_4$ injected subcutaneously into mice has reportedly resulted in formation of a malignant fibrosarcoma in the region of injection.[34] The probability of a fibrosarcoma being produced in man by subcutaneous introduction of similar amounts of plutonium is not known. Plutonium metal fragments (ranging in size from 0.67 to 1.8 mg), implanted subcutaneously into rabbits and rats;[19] did not produce fibrosarcoma. The metal was rapidly oxidized and the oxide progressively confined by the formation of collagenous connective tissue and calcification around the implantation site. From these studies, it may be concluded that plutonium metal and its oxides are relatively inert locally. One animal died, however, from an osteogenic sarcoma of the spine produced by skeletal deposition of plutonium absorbed from the implant.

The possibility of systemic absorption and local damage from plutonium embedded in the tissues necessitates industrial medical surveillance of potentially contaminated wounds. Over 50 per cent of particulate plutonium oxide contamination is removed from shallow abrasion-type wounds by normal surgical cleansing, and the rest is occluded in the scab and lost when the scab is detached. The extent of Pu^{239} contamination of lacerations and puncture wounds can be determined by external measurement of the 17 keV X-rays using a sodium iodide crystal spectrometer[35] and, at the discretion of the medical authorities, the contamination removed by surgical excision. Excision within a few days after injury can result in removal of most of the locally implanted plutonium.

Plutonium-239 *in the gastrointestinal tract.* Since the wall of the gastrointestinal tract has no cornified epithelium, Pu^{239} that is swallowed can produce α-irradiation of the mucosa during the 24–36 hr required to pass through the digestive system. Rats given 400 μg of Pu^{4+} in their drinking water for 5 consecutive days (total 2 mg) showed no signs of gross damage to the gut mucosa.[5] SULLIVAN and THOMPSON[36] administered Pu^{239} via stomach tube to approximately 200 g rats in doses of from 56 to 100 mc/kg of body weight (170 to 300 mg per animal) to see whether death could be produced and if so, whether the signs were similar to the intestinal radiation syndrome produced by irradiating the exteriorized gut with high doses of X-rays. The animals that received the lowest dose level showed no signs of damage. One out of four animals that received approximately 250 mg (88 mc/kg) of Pu^{239} died within 24 hr. Although the α-radiation dose to the intestine was estimated at about 650,000 rems, the mode of death showed no resemblance to the intestinal syndrome produced by large doses of X-rays. Lack of any signs of the acute radiation syndrome in the above experiments suggests little or no α-radiation of the radiosensitive basal cells of the gut mucosa, which in turn suggests little probability of radiation damage following chronic low level Pu^{239} ingestion.

Plutonium-239 *in the lungs.* The high incidence of lung cancer (predominantly bronchogenic carcinoma) among workers in mining operations in the Schneeberg and Joachimsthal districts of southeastern Europe was noted over 400 years ago.[37] Although the etiology of the miners' disease is still open to question, it is generally believed to have been associated with long inhalation exposure to the high concentrations of radon (3×10^{-6} $\mu c/cm^3$) and its daughters in the air of the mines.[38] LORENZ *et al.*[39] found a 50 per cent increase in pulmonary adenomas in mice after 9½ months of exposure to 8.8 r/day (total dose about 2400 r) of γ-radiation. Not only was the radiation dose relatively high and

the incidence of pulmonary tumors relatively low, but lung adenoma is very common in many mouse strains and has little resemblance to the tumors seen in the Schneeberg and Joachimsthal miners.

TEMPLE *et al.*[40] reported the effect of intratracheally injected $Pu^{239}O_2$ (using "pluronics" as a suspending agent) on the incidence of benign pulmonary papillary cystadenoma in BAF_1 mice. They found 33 per cent of the animals that received only 0.0033 μc of Pu^{239} developed lung tumors. The spontaneous incidence of papillary cystadenoma in untreated BAF_1 mice, however, was 17 per cent; the incidence in animals treated with the suspending agent only, with nonradioactive ruthenium oxide, and with 1225 r of whole body X-irradiation was 19, 28 and 62 per cent, respectively. While such observations give interesting relative information, they may not provide quantitative data on the absolute carcinogenicity of Pu^{239} particulates in the lung. These same authors, however, did find two malignant squamous cell carcinomas in BAF_1 mice 400 days after intratracheal injection of 0.006 μc of plutonium as $Pu^{239}O_2$ and one fibrosarcoma 500 days after administration of only 0.003 μc.

Production of bronchogenic cancer and other changes in the lungs of rats by β-radiation have been demonstrated with large doses (3.2 to 200 μc per animal) of Ce^{144} introduced into the lungs by tracheal intubation.[27] Introduction of lung tumors in experimental animals by other radioactive materials (Po^{210}, $BaS^{35}O_4$, Ru^{106}-oxide and Sr^{90}) have been observed also.[27] In almost all cases, the estimated radiation doses to the lungs were about 2000 rads or greater. These observations show that lung cancer can be produced by ionizing radiations and that the lungs are only moderately radiosensitive.

Plutonium deposited on the lung parenchyma in insoluble form may eventually find its way into the lymph nodes, where it may remain indefinitely.[25,26] Under conditions of long-term chronic inhalation exposure, it is possible that the pulmonary nodes may receive a greater α-radiation dose than the bone. Although there has been little observation of specific malignancy associated with lymph node accumulation of radioactive material, its potential hazard cannot be evaluated at present. The possibility of lymph node accumulation and the question of radiosensitivity of lymphoid tissue certainly suggest the need for more research on this particular aspect of plutonium radiotoxicology.

The relatively high rate of plutonium absorption from the lung (from 1 to 10 per cent of the amount inhaled), its low maximum permissible systemic burden (0.04 μc), and its 200 year biological half-time necessitate rigorous control of air concentrations in the working environment. If the lung absorption rate is indeed from 1 to 10 per cent, then the lifetime exposure of the respiratory system can be only from 0.4 to 4.0 μc. This rather strenuous limitation on lifetime inhalation exposure affords considerable protection against the potential risk of lung disease from plutonium processing.

The rigorous control of plutonium air concentrations in the working environment is largely responsible for the expensive and elaborate closed-system processing techniques that are now standard throughout the plutonium processing industry.

Skeletal deposition

Once in the blood stream, Pu^{239} is rapidly deposited in the tissues, predominantly in the bone and the liver.[5] The skeletal system is usually considered the critical organ, since it accumulates the majority of the activity and retains it essentially throughout the lifetime of the individual. Large amounts of Pu^{239} deposited in the bone and liver can produce acute or immediate radiotoxicological effects. Much smaller amounts may result many years later in bone cancer, chronic anemia, osteoporosis and bone necrosis (which may result in spontaneous bone fractures), and other symptoms seen in cases of chronic radium poisoning.

Acute effects. Relatively large amounts of Pu^{239} (500 μg/kg body weight) injected intravenously into rats produced an anemia that lasted about 90 days and an abnormally low white blood cell count that persisted throughout life.[41] Doses of 1 mg/kg body weight produced death in 50 per cent of the animals within 30 days after injection. In most cases, death occurred in from 9 to 14 days with signs

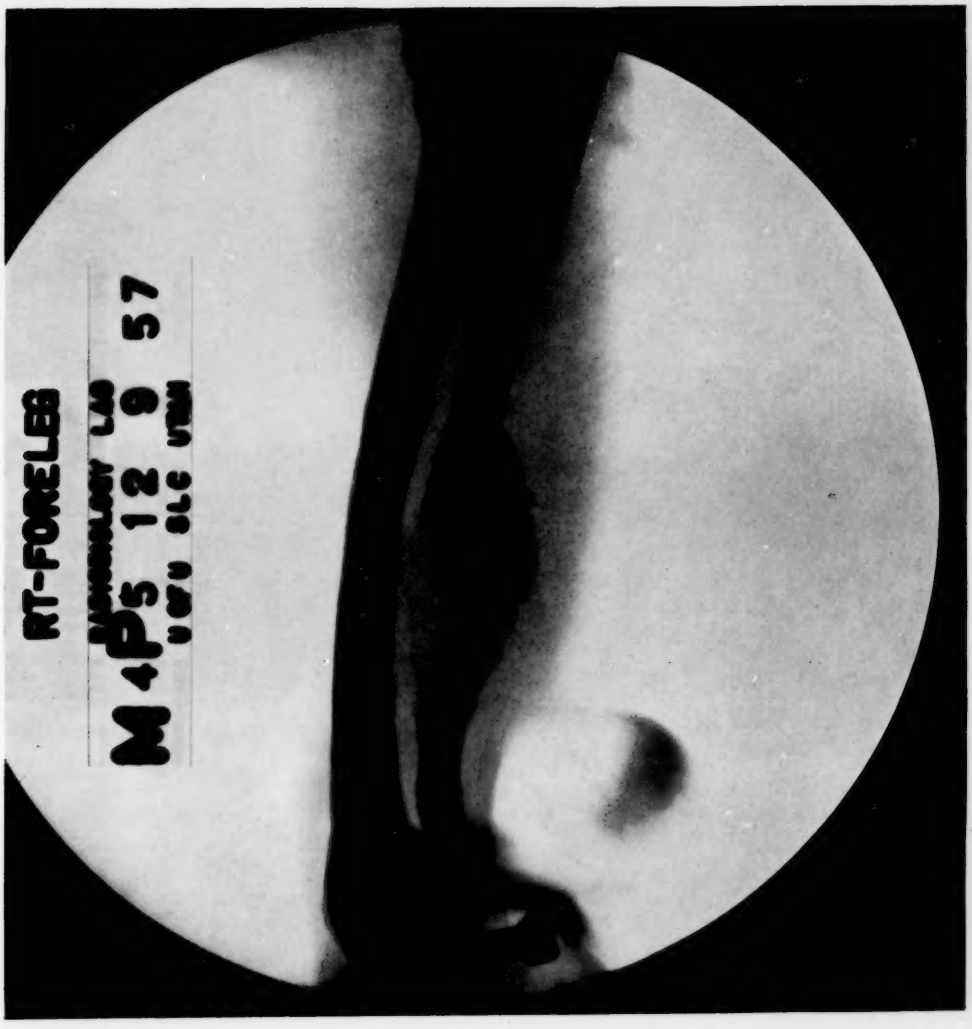

FIG. 3. Osteogenic sarcoma in a dog approximately 4 years after a retained dose of 2.5 μc Pu^{239} per kg body weight.

(*Courtesy of W. R. Christensen and C. E. Rehfeld, Radiobiology Laboratory, University of Utah.*)

comparable to those seen after an LD_{50}^{30} dose of whole body X-irradiation. The animals showed diarrhea, small areas of internal bleeding, loss of appetite, atrophy of the spleen, essentially complete destruction of the bone marrow and a disappearance of white blood cells.

Acute Pu^{239} toxicity studies in dogs showed essentially the same radiotoxicological signs and an LD_{50}^{30} of about 0.3 mg/kg. Beagle hounds given 0.27 μc/kg of Pu^{239} (equivalent to 300 μg in a 70 kg man) showed a drop in white blood cell count within 30 days, followed by a rapid return to a low normal count. A dose of 2.5 μc/kg produced an acute decrement in erythrocytes, leukocytes, heterophils and platelets with little tendency to return to normal.[42]

There is little doubt but that the signs of acute plutonium poisoning in man would be similar to those in the rat and the dog. Assuming man would respond to a weight basis like the dog or the rat, introduction of from 20 to 70 mg of Pu^{239} into the systemic circulation would result in a 50 per cent chance of death within 30 days. An individual surviving beyond 30 days would have an extremely poor prognosis and would surely succumb later to the chronic or delayed effects of such an overwhelming dose. The median survival time of rats that survived an acute LD_{50}^{30} was about 40 per cent of that of the controls. While the results of such a systemic exposure would surely be catastrophic, its probability of occurrence is extremely slight. With a gastrointestinal absorption rate of 0.003 per cent, an individual would have to ingest about $1\frac{1}{2}$ lb of plutonium. With a lung absorption rate of from 1 to 10 per cent, one would have to inhale from 0.2 to 2 g in the form of particles or droplets of a few microns or less in diameter. An explosive-type accident involving concentrated plutonium solutions and serious traumatic injury could conceivably occur in which lethal amounts of material could be deposited in the peritoneal cavity and absorbed.

Chronic or delayed effects. Animal experiments have shown beyond doubt that deposition in the skeleton of amounts of Pu^{239} too small to produce signs of acute damage may eventually produce serious bone pathology, including osteogenic sarcoma (Fig. 3). The latent period between exposure and appearance of damage is about 25 to 50 per cent of the normal life expectancy of the species. The induction period between time of exposure and time of appearance of tumors in eight cases of radium-induced malignancy in humans reported by Aub *et al.*[1] was 12 to 30 years, with an average of 23. These data and those reported by others[2–4] show that from 0.7 to 1 μc of Ra^{226} fixed in the skeleton for 25 years or longer may produce significant bone disease, and 0.8 μc has produced osteogenic sarcoma. Since information on the chronic or delayed radiotoxicity of Ra^{226} in man is available, the radiotoxicity of Pu^{239} is estimated from its tumorigenic potency relative to radium when administered to experimental animals.

Finkel[43] studied the relative potency of intravenously injected Ra^{226} and Pu^{239} for production of bone tumors in mice (Fig. 4).

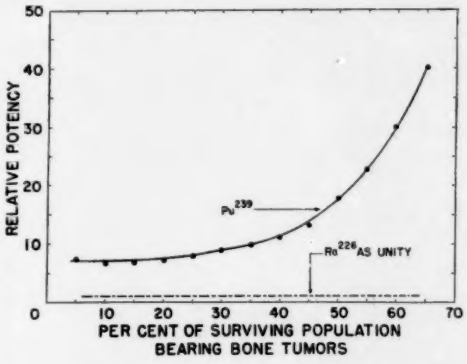

Fig. 4. Relative tumorigenic potency of Ra^{226} and Pu^{239} administered intravenously to mice.[43]

These data showed that plutonium was about thirty times as potent as radium when large enough doses were given to produce tumors in 60 per cent of the animal population. As the dose (and consequently the tumor incidence) decreased, plutonium showed a plateau in relative tumorigenic effect at about seven times that of radium. In these studies (using over 1400 mice), comparison was made on the basis of the dose injected. When the relative potency was compared on the basis of radiation dose to the skeleton (by allowing for relative skeletal retention of Pu^{239} and Ra^{226}, and the exhalation

of radon by the mouse), Pu^{239} was about four times as tumorigenic as Ra^{226}.

About 8 years ago, the AEC established a project at the University of Utah to study the relative radiotoxicity of Ra^{226}, Pu^{239}, MsTh(Ra^{228}) and RdTh(Th^{228}) in mature beagle hounds. The injected doses of the various nuclides were such that the amounts (in μc/kg body weight) retained by the animals were equivalent to from 10 to 1600 times the presently accepted maximum permissible body burden (0.0014 μc/kg) of Ra^{226} for man. Some of these animals have carried their body burdens for about 6 years, and extensive bone pathology (including bone tumors) has occurred in the high dosage groups.[44] Four animals that retained the Pu^{239} equivalent of 180 times the human maximum permissible Ra^{226} level have died of bone tumors. No tumors have yet developed in the dosage group that is sixty times the human maximum permissible level. These observations cannot be used at present as an indication of the absolute Pu^{239} tumorigenic dose, since the experiment still has several years to go and tumors may occur yet in the lower dosage levels. The data at the present time do indicate, however, that Pu^{239} indeed may be about five times as hazardous (on the basis of equivalent radiation dose to the skeleton) as Ra^{226}.

The obvious industrial medical regulation of the potential hazards of internally-deposited Pu^{219} is to control all its potential routes of entry into the systemic circulation such that the probability of workers accumulating appreciable body burdens is small.

MAXIMUM PERMISSIBLE BODY BURDEN OF PLUTONIUM-239

The use of Ra^{226} (and MsTh) in the luminous dial industry, public consumption of radium-rich waters, and the therapeutic application of radium by the medical profession have provided information on the radiotoxicology of this material in man. On the basis of these experiences, the National and International Commissions on Radiological Protection adopted 0.1 μc as the maximum permissible burden of Ra^{226} for occupational exposure. The maximum permissible body burden may be defined as the maximum amount of material that can be maintained indefinitely in the adult human body without producing significant bodily injury to any person at any time during his natural lifetime.

Since Pu^{239} (like radium) emits most of its radiant energy as α-particles and concentrates predominantly in bone, the skeleton is considered the critical organ and the maximum permissible body burden for occupational exposure is determined by comparison with that of Ra^{226}. The maximum permissible body burden (q) may be estimated by the following expression:

$$q = \frac{q_{Ra} \times f_{Ra} \times E_{Ra}}{f_{Pu} \times E_{Pu} \times 5}$$

$$= \frac{0.1 \times 1 \times 10.4}{0.9 \times 5.3 \times 5} = 0.043 \ \mu c$$

in which q_{Ra} is the maximum permissible body burden of Ra^{226}, f_{Ra} and f_{Pu} are the respective fractions of total body radium and plutonium deposited in the skeleton, E_{Ra} is the energy (in MeV) deposited in tissue per Ra^{226} disintegration (taking into consideration recoil energy, daughter decays and fractional loss of radon by exhalation), E_{Pu} is the energy per Pu^{239} disintegration (including recoil energy), and 5 is the radiotoxicological potency of Pu^{239} relative to Ra^{226} as determined by animal experiments. On this basis, 0.04 μc is the maximum amount of Pu^{239} which, when fixed indefinitely in the human body, has the same improbability of producing significant bodily injury as does 0.1 μc of Ra^{226}.

ESTIMATION OF PLUTONIUM-239 BODY BURDEN

An individual's Pu^{239} body burden may be determined from a 24 hr urine analysis[25] and the urinary excretion expression given as equation (1). Following a single acute exposure occurring at known time, the body burden (D_E) at time of exposure is given by the expression:

$$D_E = 500 U t^{0.74} \qquad (4)$$

where U is the amount of Pu^{239} found in a 24 hr urine sample collected t days after exposure. The body burden at time of exposure is given in whatever units (c/m, d/m, μc or μg)

are used to express U. The retained body burden (D_R) at time t following a single acute exposure is given by the expression:

$$D_R = 435Ut^{0.76}. \quad (5)$$

The exposure dose received by an individual as a result of chronic variable exposure of known duration (i.e. the time worked since the last negative urine assay) may be approximated from the assay of a 24 hr urine specimen and the same expression used for acute exposure occurring at known time (equation (4)). One may assume that the individual obtained all his body burden on the first day of exposure, in which case t becomes the elapsed time from beginning of work to the time of collection of the urine sample. Unless the individual actually did accumulate his body burden on the first day of work, such an estimate will be too high. One may assume also that the body burden was obtained on the last day of work, in which case t becomes the elapsed time between the last day of work and the time of collection of the urine specimen. In this case, the estimate may be too low. One may also average the results obtained on the basis of the two assumptions made above. The average result, of course, has the greatest chance of carrying the smallest error.

Following chronic invariant exposure to Pu^{239} (as might occur under conditions where air concentrations are rigidly controlled, the work highly routine, and the material uniformly distributed throughout the working environment), the total body exposure (T_D) may be calculated from the expression:

$$T_D = \frac{130 \times m \times U_n}{(n + \frac{1}{2})^{0.26} - (n - m + \frac{1}{2})^{0.26}} \quad (6)$$

where m is the duration of exposure (in days), n is the time from beginning of exposure until urine sample was taken, and U_n is the amount of Pu^{239} found in the 24 hr urine sample. Although the above expression is derived from the basic urinary excretion equation, it probably has little practical application to plutonium processing, where the materials are usually not distributed uniformly throughout the working environment.

Because of fluctuations in urinary excretion and statistical variations in the method of analysis, plutonium body burden (in actual practice) is not determined from a single urine sample but from a series of samples.

Several methods for determination of Pu^{239} in urine have been devised. The most sensitive procedure is the one developed by SCHWENDIMAN *et al.*[45] The urine sample is wet ashed with nitric acid and the plutonium coprecipitated with lanthanum fluoride. After further separation by extraction with thenoyltrifluoroacetone in benzene, the plutonium is converted to PuO_2^{2+} and electrodeposited on a stainless steel disk. The disk is placed in contact with a nuclear track α-plate for approximately 1 week, and the number of tracks registered on the developed plate are counted visually with a microscope. Plutonium recoveries of 85 ± 5 per cent are obtained routinely, and the detection limit at the 99 per cent confidence level is about 0.05 d/m per 24 hr urine sample (3.6×10^{-13} g of Pu^{239}). Although the above method is the one of choice for estimation of small plutonium body burdens, it is extremely tedious and time-consuming and several days are required to obtain a result. In an emergency involving potentially high exposure, a more rapid, less sensitive, method is highly desirable. A satisfactory procedure consists of digesting a 200 ml urine sample with HNO_3, followed by two lanthanum fluoride coprecipitations. The second precipitate is dissolved in aluminum nitrate solution and the plutonium extracted with thenoyltrifluoroacetone in benzene. The extract is evaporated in a counting dish and counted in a proportional α-counter.[46] The result can be obtained in about 2 hr after collection of the sample.

ACCELERATION OF PLUTONIUM-239 EXCRETION

Numerous attempts to remove Pu^{239} from the animal body have been made. Three substances that have shown promise are ethylenediaminetetra-acetic acid,[47] diethylenetriaminepentaacetic acid[48] and zirconium citrate.[49] The first two are strong complexing agents that mobilize plutonium into the blood stream and accelerate its excretion via the kidneys.

Zirconium citrate is believed to exert its effect through a colloidal ion exchange process in which the final disposition of plutonium follows that of the zirconium colloid. All three treatments are most effective if applied immediately after exposure before the plutonium has become fixed in the bone. Diethylenetriaminepenta-acetic acid looks promising in animal experiments but has not been tried in humans. Rats treated 1 hr after intravenous Pu^{239} injection showed 9 per cent fixation of plutonium in the skeleton, compared to 61 per cent for untreated controls. Animals treated several times per week for 4 weeks, beginning 38 days after plutonium injection, had about half the skeletal burden of untreated controls.[48] Rats treated with zirconium citrate at 1 hr after plutonium injection showed 20 per cent deposition in the skeleton, compared to 68 per cent in untreated controls.[49] Treatment of human exposure cases, however, has met with little success.[50]

FOREMAN *et al.*[51] reported treatment of two Pu^{239} exposure cases with ethylenediaminetetra-acetate. In one case, treatment was begun 5 days after exposure and produced a decrease of about 25 per cent in the individual's body burden. Better results might have been obtained had treatment been started earlier. Treatment of the second individual, who had accumulated plutonium through long continued chronic exposure, failed to produce a significant change in the body burden. Ethylenediaminetetra-acetic acid has been shown to produce kidney damage.[52] Since kidney damage from ethylenediaminetetra-acetate is believed to be related to its strong complexing action for divalent ions, diethylenetriaminepenta-acetate may be nephrotoxic also.

The decision to treat or not to treat a case of plutonium exposure is difficult. Each case must be decided individually and, since the methods of treatment become less effective with time, the decision should be made as soon as possible. One microcurie of Ra^{226} (ten times the maximum permissible body burden) has produced serious bone disease in a few individuals. Ten times the maximum permissible body burden of Pu^{239} (0.4 μc) might be expected to produce severe bone changes in a few individuals also. The treatment of cases with several times the maximum permissible body burden would certainly seem justified. Indiscriminate treatment, however, should be avoided in view of the potential toxicity of the drugs.

HUMAN EXPERIENCE WITH PLUTONIUM-239 EXPOSURE

Occupational exposures to Pu^{239} have occurred at the Los Alamos Scientific Laboratory, General Electric's Hanford Atomic Products Operation (Richland, Washington), Canadian Atomic Energy Agency (Chalk River), and the British Atomic Authority.

PARKER[53] reported the plutonium exposure history of 10 years of operation of the General Electric's Hanford facilities. One hundred cases of positive plutonium exposure have been detected. There has not been a single case of overexposure, and in only two cases was the body burden greater than 50 per cent of the maximum permissible level. In 70 per cent of the cases, the exposures were 10 per cent or less of the maximum permissible body burden. Fifty-five per cent of the cases were associated with known exposure incidents and 45 per cent with chronic exposure under apparently normal working conditions. Of the cases associated with known exposure incidents, 75 per cent were exposed via inhalation. This supports the view long held at Los Alamos that absorption through the lungs is the major potential route of entry of plutonium into the systemic circulation.

The Canadian Atomic Energy Establishment at Chalk River had an accident in 1950 in which two individuals received plutonium exposure. One individual received a body burden of 0.15 μg (0.01 μc) and the other 0.10μg (0.006 μc). CIPRIANI[54] reported also that thirty-one out of 321 Chalk River employees tested during 1953–1954 had detectable Pu^{239} body burdens. The British Atomic Energy Authority had a contaminated accident at Harwell in 1952 in which a waste disposal worker received a body burden of approximately 5 μg (0.32 μc). The mode of exposure was not known, but it was probably via inhalation.

The oldest exposure cases on record are those reported by the Los Alamos Scientific Laboratory.[55] Most of these occurred from the fall of 1944 through 1945. During this

period, twenty-seven workers accumulated Pu^{239} body burdens of 0.1 μg or greater. Eleven accumulated levels equal to or greater than the presently accepted maximum permissible body burden, three of which were about twice that amount. Most of the exposures were believed to have occurred via inhalation as evidenced by the strong correlation with frequent contamination of the nasal vestibule determined by counting nasal swabs rotated in the nares immediately following highly contaminated operations.

The air-borne activity to which some of these persons were exposed was occasionally orders of magnitude above presently accepted maximum permissible air concentrations. On one occasion, the nasal swabs from an individual yielded over 1 μg of plutonium from each nostril. His body burden after this and several other similar operations was only 0.5 to 1 μg.

Twenty-four of the twenty-seven Los Alamos cases are being followed routinely at 3 year intervals for any signs of chronic or delayed effects. Complete general physical examinations, including laboratory tests, hematological studies, and X-rays, are conducted. Roentgenograms include pelvis, chest, skull, knee, elbow, hand and jaw. The roentgenograms are studied for signs attributable to plutonium. At the 9 year period, all observations were negative. When contacted in preparation for the 12 year follow-up examination, all subjects reported a continued state of normal health. Although the critical period for appearance of chronic effects has not passed, the complete negative character of the observations is encouraging.

DISCUSSION

Because of the superior fission properties of Pu^{239}, the future of the plutonium processing industry and the plutonium specialist is assured. However, because of the potential hazard of plutonium, assured also is the future of the industrial hygienist, the safety engineer and the industrial physician. What then is the situation with regard to the necessity for elaborate industrial engineering control of processes involving large amounts of this material? Management's prime consideration, of course, must be the health of the employee, since relatively small amounts of plutonium taken into the body and deposited in the skeleton may may predispose the individual to serious bone disease many years later. The maximum permissible body burden (0.04 μc) is considered safe in this respect. There are, however, important secondary factors that management must consider also. These may be illustrated by means of the following hypothetical example.

Suppose a young metallurgist, "John Doe", at the age of 21 accepts a position with a hypothetical company, "Plutonium Rocket Motors, Inc." Without adequate knowledge of the basic physiology and toxicology of plutonium, the first thing that is apt to happen to Mr. Doe (when he sees the elaborate industrial engineering control and persistent monitoring for air and surface contamination, is asked to wear protective clothing and rubber gloves, keep a respirator always at hand, check his hand contamination frequently, and submit routine urine samples) is an unwarranted fear for his personal health and safety. His apprehension could result in his creating a greater potential hazard to himself and those about him.

Let us assume also that in a few years Mr. Doe has become a highly skilled specialist in alloying, casting and welding plutonium metal, but in the process has accumulated a maximum permissible body burden. Although there is no reason to feel that his physical well-being has been jeopardized, he will still have 80 to 90 per cent of his body burden at retirement age and technically should be removed from further contact with plutonium for the remainder of his working lifetime. In this case, the company has lost the technical skill of an important specialist.

Furthermore, the question arises as to what the management can do with Mr. Doe. The problem of removing an employee from his job, in the interest of his health or well being, is not unique to the plutonium industry. Other industries have faced similar dilemmas. The alternatives are as follows: (1) The company can discharge Mr. Doe, which most certainly will do little to further labor–management relations. Moreover, Mr. Doe may choose to test the legality of the action. (2) Management can transfer Mr. Doe (the action taken usually by other industries) to other technical work not involving plutonium, or promote him to an

administrative or supervisory position outside the processing area. In this case, there is the possibility that (a) a satisfactory job, involving no potential plutonium exposure, may not be available, or (b) Mr. Doe may not be qualified or may not wish to be an administrator or a supervisor. (3) The company can seek a settlement with Mr. Doe through its own devices or through workers' compensation laws, few (if any) of which specifically cover the particular type of claim.

Because of conscientious industrial medical and engineering control over all plutonium operations, management has rarely faced the situation of Mr. John Doe. The cases of maximum permissible exposure that occurred at the Los Alamos Scientific Laboratory occurred during and immediately after the War and were mostly young college graduates assigned to the Laboratory through the Manhattan District. Upon discharge, some returned to jobs unassociated with atomic energy development. Others remained at the Laboratory where, by virtue of their technical knowledge and ability, they progressed naturally to key administrative and supervisory positions. This fortunate circumstance, however, should not be expected to occur always.

In addition to continued rigorous industrial hygiene and engineering control of all plutonium operations, there are a few courses of action which seem to merit industry's future consideration. First, existing workers' compensation laws may be modified and new ones written to deal specifically with the industrial risks of the nuclear energy development program. Second, company insurance plans may be developed that protect both employee and employer in the event of a lifetime maximum permissible exposure. Third, the lifetime maximum permissible Pu^{239} exposure may be prorated against the employee's potential employment expectancy as is now customary for lifetime exposure to whole body irradiation. In this case, an employee's maximum permissible body burden should be:

$$\frac{0.043}{47}(N - 18) = \sim 0.001(N - 18)\ \mu c$$

in which 18 is the legal employment age, 47 is the number of years of expected employment before retirement at age 65, and N is the chronologic age of the individual. There are a number of interesting facets to this proposal. (1) An individual's maximum permissible body burden is dependent only on his chronologic age and not on his length of employment. (2) Other things being equal, older people are favored as routine plutonium process operators. (3) An additional safety factor is provided in that an individual is not allowed his lifetime maximum permissible body burden until retirement age. (4) It is somewhat conservative in that it assumes an individual, once he is employed, will work with plutonium for the rest of his occupational lifetime.

Insistence on rigorous industrial engineering control does not mean that accumulation of a Pu^{239} maximum permissible body burden has any great probability of producing significant bodily injury. It means primarily that plutonium processing without such control would be contrary to good industrial hygiene and to the elementary principles of industrial management. If presently recommended industrial medical practices are maintained, there is little reason to feel that the health of an individual working with plutonium will be subject to any greater absolute risk than if he were driving a truck, operating a lathe, or working in an ordinary chemistry laboratory.

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RAPID DETERMINATION OF MIXED BETA–GAMMA RADIONUCLIDES IN URINE*

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Abstract—A bio-assay procedure was developed which enables a rapid estimation of the total amount of β–γ emitting isotopes in a urine specimen. The β–γ emitting radionuclides are coprecipitated from urine on an ammoniacal alkaline earth phosphate precipitate. The filtered precipitate is fired in a muffle furnace, dissolved, and directly plancheted. The planchet is both β- and γ-counted. The recoveries of strontium-89, strontium–yttrium-90, zirconium–niobium-95, cerium–praseodymium-144, iron-59, chromium-51, and zinc-65 are greater than 90 per cent. Cobalt has a recovery of 85 per cent.

INTRODUCTION

THE routine analysis of urine samples as a quick check for the possible assimilation of fission products and induced activities by operating personnel is complicated by the large number of β–γ emitting radionuclides encountered. Because of the large amount of time required for their individual analysis, a single method for the recovery of all the radionuclides of interest would be advantageous. One group of radionuclides of concern, commonly known as fission products, are isotopes of strontium, yttrium, rare earths, ruthenium, zirconium and niobium. Other radionuclides of concern include isotopes of iron, cobalt, chromium and zinc, made radioactive by neutron irradiation of stable isotopes and referred to in this report as induced activities. Attempts to recover collectively all of these radionuclides on a single precipitate culminated in the use of an ammoniacal alkaline earth phosphate precipitation. A phosphate method was previously reported[1] for barium, strontium and rare earths, but this method was not easily applied to large numbers of routine samples. Preliminary investigation indicated that the ammoniacal alkaline earth phosphate method was more applicable to total β–γ determinations and it was therefore evaluated for this purpose.

PROCEDURE

Sample preparation. To a 750 ml urine sample in a 1000 ml beaker, add 5 ml of concentrated nitric acid, 2 ml of 85.8% orthophosphoric acid, and 2 ml of a 25 mg/ml cobalt carrier solution.* Heat the sample to 85°C. Slowly add 25 ml of concentrated ammonium hydroxide with continuous stirring. Continue to stir and heat the solution at 85°C for 2 hr. Cover the beaker with a watchglass and let the precipitate settle overnight. Decant the supernate, being careful not to disturb the precipitate. Using a "Fisher Filtrator" funnel and a suction flask, collect the precipitate on a Whatman no. 50 filter paper. Quantitatively transfer the remaining precipitate to the funnel with distilled water. Put the filter in a 30 ml porcelain crucible and place the crucible in a cold muffle furnace. Fire the precipitate at 900°C until it is completely white; cool and, with heat, dissolve the residue in 10 ml of concentrated nitric acid. The addition of more nitric acid will aid in dissolving when the initial amount has partially evaporated. Quantitatively transfer the solution to a 50 ml beaker. Evaporate the solution to 2 ml and using 8 N nitric acid quantitatively transfer to a flamed stainless steel planchet. Add all washes to the planchet, evaporate to dryness and count.

* The information contained in this paper was developed during the course of work under Contract AT(07-2)-1 with the Atomic Energy Commission.

* All chemicals used are of reagent grade and no further purification is necessary. The cobalt carrier solution is made by dissolving 25 g of cobaltous chloride ($CoCl_2 \cdot 6H_2O$) in 250 ml of 0.01 N HCl.

β- and γ-counting. Determine the β-activity of the sample by counting the emitted β-rays with a G.M. or proportional counter. Cover the planchet evenly with cellophane tape and determine the γ-activity by counting the sample with a sodium iodide crystal scintillation counter.

DISCUSSION

The ammoniacal phosphate precipitation utilizes the calcium already present in raw urine to form a general scavenging precipitate. Upon the addition of concentrated ammonium hydroxide and phosphoric acid, the induced activities and the fission products present in the urine will form either basic phosphates or hydroxides. In every case studied concerning the formation of a hydroxide precipitate rather than the basic phosphate precipitate, an excess of ammonia is avoided, owing to the solubility of the hydroxides of the divalent elements. Because of this effect, cobalt carrier was added to increase the recovery of radioactive cobalt (Table 1). The solution is heated and stirred for 2 hr to obtain complete coprecipitation. The precipitate is fired in a muffle furnace to eliminate organic matter. The amount of precipitate collected on the planchet from a 750 ml specimen is approximately 1 g and is easily mounted.*

The β-count is primarily used to determine the presence of strontium-89, strontium-90 and yttrium-90 which emit no γ-radiation. Because high energy β-rays are associated with strontium-89, a reliable measurement of its activity is made even though an average of 1 g of precipitate is mounted on the planchet. Yttrium-90, which emits a strong β-ray, also yields a reliable count, and affords a satisfactory estimate of its parent isotope, strontium-90, since some strontium-90 β-rays are not counted, owing to absorption by the solid. The low energy β-rays of most of the induced activities along with the β-rays of zirconium–niobium and the rare earths also contribute to the total gross β-ray count. No interference from potassium-40 is observed.

* A volume of 750 ml of urine was selected since from it could be obtained the desired sensitivity. Larger or smaller volumes of urine affect only the sensitivity of the method.

Table 1. *Recovery of radionuclides added to* 750 ml *of urine*

Nuclide	Recovery (%)	Standard deviation (±%)
Induced activities		
Co-60*	40	—
Co-60†	85	6
Fe-59	98	7
Zn-65	95	5
Cr-51	90	8
Fission products		
Sr-89‡	92	6
Ce, Pr-144	101	6
Sr, Y-90§	103	9
Zr, Nb-95	95	3
Ru-103	40 to 50	—

* Without carrier.
† With 25 mg of carrier.
‡ No correction for absorption.
§ Based on yttrium count.

The γ-count is primarily used to determine the presence of induced activity. A γ-count is used because of the low counting efficiency of the weak β-energies associated with the induced activities. The γ-ray from zirconium–niobium

Table 2. *Procedure sensitivity*

Nuclide	Lower limit of sensitivity per 750 ml of samples (μc)
Sr, Y-90	1.3×10^{-5}*
Sr-89	0.34×10^{-5}*
Zr, Nb-95	1.4×10^{-4}†
Nb-95	1.8×10^{-4}†
Ce, Pr-144	1.9×10^{-5}*
Co-60	1.5×10^{-4}†
Fe-59	2.2×10^{-4}†
Cr-51	10.0×10^{-4}†
Zn-65	6.8×10^{-4}†

* Based on β-count.
† Based on γ-count.

Table 3. Chemical characteristics of tracer solutions

Isotope	Received as*	Used as
Ce, Pr-144	$CeCl_3$ in HCl	Ce^{III}
Co-60	$CoCl_2$ in HCl	Co^{II}
Cr-51	$CrCl_3$ in HCl	Cr^{III}
Fe-59	$FeCl_3$ in HCl	Fe^{III}
Ru-103	$RuCl_3$ in HCl	Ru^{III}
Sr-89	$SrCl_2$ in HCl	Sr^{II}
Sr, Y-90	$SrCl_2$ in HCl	Sr^{II}
Zn-65	$ZnCl_2$ in HCl	Zn^{II}
Zr, Nb-95	Complex in oxalic acid	Zr^{IV}

* All radioisotopes were obtained from Isotopes Sales Department, Oak Ridge National Laboratory. Solutions were diluted with 1 N HCl or HNO_3.

and rare earths also contributes to the total gross γ-count.

Recoveries of the individual β–γ nuclides studied are greater than 85 per cent with the exception of ruthenium (Table 1). The lower limits of sensitivity for the individual radionuclides, based on the β- and γ-counting statistics at the 90 per cent confidence level are listed in Table 2. The chemical forms of the tracers used are listed in Table 3.

Specific chemical analyses for the individual radionuclides can easily be made on a positive precipitate without loss of time due to resampling. A γ-ray pulse height analysis of a positive sample can also be used to distinguish between the different γ-emitting nuclides present as a quick qualitative approach before specific chemical analyses are made.

The mixed β–γ isotope method provides a simple evaluation of the sample regarding the need for an extensive specific analysis, thus eliminating unnecessary and time consuming work.

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A DIAGNOSTIC X-RAY EXPOSURE DOSE CHAMBER*†

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Abstract—The design of a miniature condenser ionization chamber for the measurement of diagnostic X-ray exposure dose is described. Response relative to the roentgen as a function of energy for X-rays with a range of half value layers from 0.2 to 5.0 mm Al (effective energies of 12.8 and 41.5 keV, respectively) is kept constant by the correct choice of chamber dimensions and by constructing the chamber entirely of material of lower atomic number than that of air. Measured values of sensitivity and directional dependence are presented together with the calculated maximum dose rate. The chambers can be made in large numbers by a molding technique with completely reproducible properties.

ALTHOUGH a variety of radiation detectors has been developed for X-rays, no miniature detector has so far been developed which is specifically designed to measure exposure dose in the diagnostic X-ray region. Diagnostic X-rays are primarily produced by machines with peak voltages between 50 kVp and 150 kVp. Most detectors have been designed for therapeutic X-ray energies above 200 kVp. A diagnostic detector should satisfy the following criteria: be uniformly sensitive to all of the different energies of X-rays in the diagnostic region; be uniformly sensitive for different directions of the incident radiation; have a linear response which is independent of the highest dose rates employed in diagnostic procedures; have a sufficiently low leakage so that protracted exposures are feasible; and have a miniature size in order that the measurements may be made essentially at a point either in space, in a body, or in a simulated body.

Various detector systems considered for this purpose include: (a) certain semiconductors whose conductivity is radiation dependent; (b) scintillation crystal response; (c) photographic film response; (d) ionization chamber response; and (e) certain radiation chemical systems. The first, second and fourth of these systems can be used for continuous recording of exposure dose rate. The third, fourth and fifth can be used for the integration of dose at a given point. On the basis of purely technical considerations, such as reproducibility, ease of satisfying the criteria cited above, and sensitivity, the development of a condenser type miniature ionization chamber method was decided upon and is described here. Other systems for the same purpose are still in a state of development.

DESIGN PRINCIPLES IN THE DIAGNOSTIC X-RAY REGION

The first design principle which will be considered is uniformity of sensitivity as a function of X-ray energy. The maximum energy of X-rays used in the diagnostic region corresponds to a half value layer of approximately 3 mm Al (33 keV). The corresponding effective energy will be indicated in parentheses throughout this paper. Effective energy as used here is defined as the energy of the monochromatic radiation for which the indicated thickness of aluminium will reduce its intensity by a factor of 2. In order to measure scattered radiation and radiation in the useful beam, the chamber should have a response independent of energy for X-rays whose quality may be

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represented by this half value layer and all lesser half value layers down to a few-tenths of a millimeter of aluminum.

The uniformity of sensitivity should be measured relative to either the exposure dose in roentgens (r) or the absorbed dose in rads. Since exposure dose is experimentally better defined in this region, it was decided first to develop chambers which had a uniform response relative to the roentgen, which is the chamber described here. Since the roentgen is defined in terms of absorption in air, such a chamber should have air equivalent walls which provide equilibrium in X-ray absorption and electron production, without X-ray attenuation. A truly air equivalent wall is difficult to attain owing to the critical dependence of X-ray absorption on atomic number in this X-ray region. Also attenuation in the wall is a serious problem at these low energies.

Design principles on the basis of which the response of a chamber may be made constant in the diagnostic region have been discussed elsewhere.[5] The two chambers described here were constructed as follows. Chamber A was constructed entirely of material of atomic number lower than that of air. With a correct choice of chamber dimensions this method will yield a response nearly independent of energy over the diagnostic region[1] (see Fig. 1). Chamber B's collecting electrode was of atomic number greater than that of air in order to elevate the low energy response by increased production of photo-electrons.

As described by GARRETT *et al.*[5] the principle on which chamber *A* operates is as follows: with high energy X-rays, ionization within the chamber is due mainly to secondary electrons emitted from the walls. As the energy of the X-rays is reduced, the range of these secondary electrons decreases and they no longer fully traverse the chamber, so that direct ionization of the chamber gas contributes relatively more ions to the total number collected. However, the gas (air) has a higher atomic number than the wall and hence a greater cross-section for photoelectric absorption, so that the number of ions formed per second for a given exposure dose tends to increase as the energy is reduced. This increase is counteracted by wall attenuation so that by a correct choice of chamber air volume and wall thickness a constant sensitivity may be obtained over a range of half value layers from 0.2(12.8 keV) to 5.0 mm Al (41.5 keV).

Another criterion which must be satisfied is uniform directional response. Chambers are normally calibrated with their long axis normal

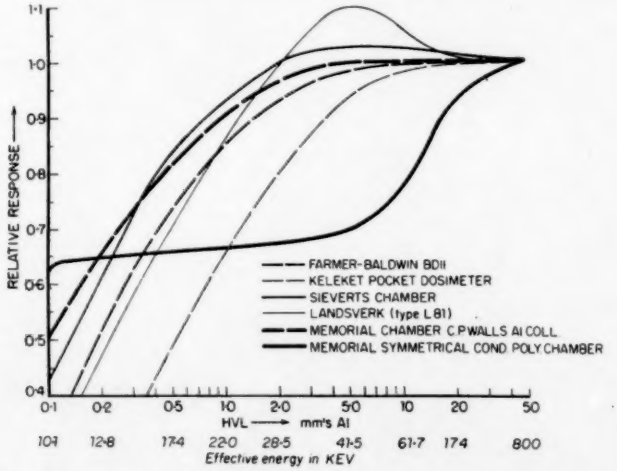

FIG. 1. General response characteristics of some other detectors and the two condenser chambers described in this paper. In this figure the actual sensitivities have been normalized at a half value layer of 50 mm Al.

to the axis of the X-radiation. The response of the chamber may be quite different in other directions. In some commercial chambers the response may vary by as much as a factor of 3, depending on the angular relation of its axis to that of the incident radiation.

A further criterion which is particularly important for the high dose rates encountered in X-ray diagnosis is that the chamber must respond linearly regardless of intensity. This requires that throughout the collecting volume of the chamber the electric field be sufficiently high so that essentially all ions be collected before recombination can occur. This can be tested by calibration of a chamber at a variety of dose rates.

DESIGN OF MEMORIAL DIAGNOSTIC CHAMBERS

The walls and collecting electrode of chamber A, Fig. 3 are made of conducting polystyrene. Insulation used on the chambers so far has been teflon. This, however, shows some leakage and future chambers are being fitted with molded polystyrene insulators.

Since the chamber sensitivity and energy response are both functions of the wall material and the chamber dimensions, development of an energy independent chamber was carried out by fixing the dimensions at suitable values (inside length 3 cm, inside diameter 1 cm). The response as a function of half value layer was then adjusted by varying the wall thickness and the collecting electrode diameter, thus varying the air volume. Dimensions which give a uniform response for half value layers between 0.2 (12.8 keV) and 5.0 mm of Al (41.5 keV) are shown in Fig. 3. Despite its small size, a condenser chamber of this design has a low capacity resulting in a sensitivity of approximately 0.4 V/mr.

Location of the insulator at the center of the chamber instead of at one end was important in obtaining a response as nearly independent of direction as possible. This arrangement allows the use of a thinner insulator and alleviates the excessive attenuation at low energies which would otherwise occur in the direction of the connector end. Connection to the collector electrode is made by removal of a small plug which has the same thickness as the wall.

Chamber B, also in Fig. 3, has been designed on principle (2) above, i.e. with an aluminum collecting electrode and a conducting polystyrene wall of 2 mm thickness, and exhibits an energy independent response over a range of half value layers from 5.0 mm Al (41.5 keV) to 2.0 mm Cu (105 keV).

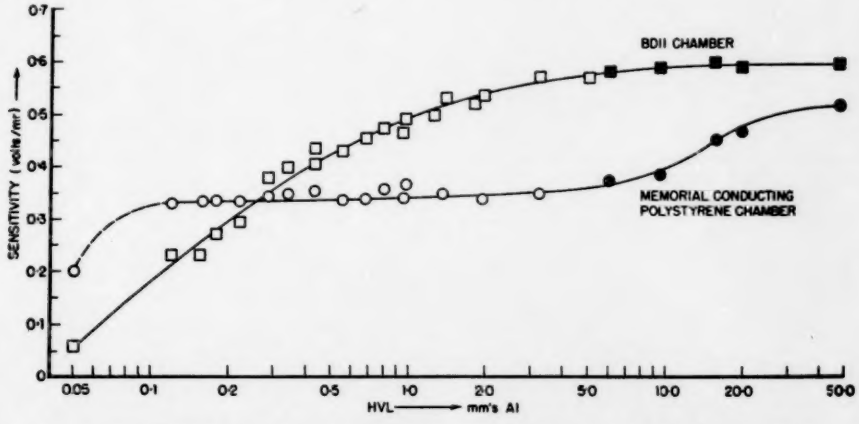

FIG. 2. Sensitivity (V/mr) as a function of the half value layer of the incident radiation. The Memorial chamber A is shown compared with the Baldwin–Farmer type BD11 chamber. The shaded points are those obtained with a constant potential X-ray machine as radiation source. The other points were obtained with an OEG60 X-ray tube operated with a half-wave rectified supply.

PERFORMANCE DATA

In all the measurements described here the condenser chambers have been used in conjunction with a Baldwin–Farmer electrometer.[2] With this instrument the chambers are charged initially to 250 V and the decrease in voltage due to irradiation is measured by means of a low input capacitance electrometer tube circuit.[2]

Energy response measurement for half value layers less than 1.8 mm Al were made with a Machlet type OEG60 tube, operated at various voltages from 60 kV down, supplied by a half wave rectified power supply. A thin window Victoreen chamber type 651 was used as a

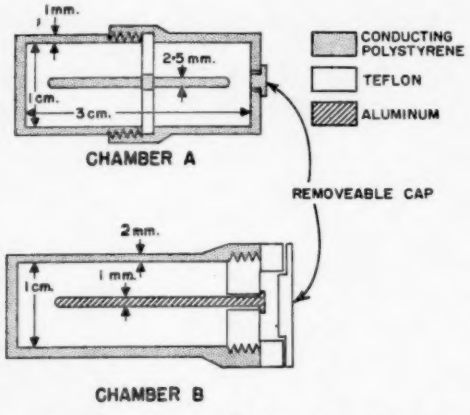

FIG. 3. Dimensions and materials of the conducting polystyrene ionization chambers described in the text.

standard. This Victoreen chamber was calibrated at the Bureau of Standards for half value layers between 0.05 (8 keV) and 1.0 mm Al (22 keV).

Both the chamber being calibrated and the Victoreen standard, were exposed at the same point in the X-ray beam, but for different times, depending on their sensitivities. The miniature chambers required around 20 sec exposure time whereas the Victoreen required about 1 hr.

For measurements at half value layers of 1.8 mm Al (27.5 keV) and greater, a Machlet 250 kV beryllium window tube was used in conjunction with a constant potential generator which provided continuously variable voltages from 60 kV to 250 kV. Actual calibrations were made against a 25 r bakelite Victoreen

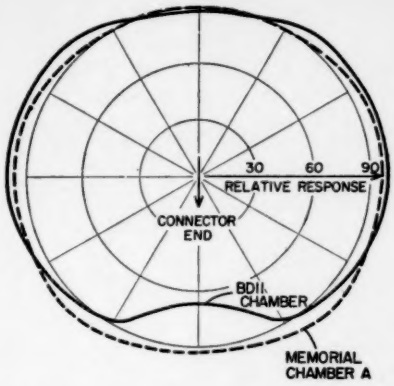

FIG. 4. Directional response of the miniature cylindrical ionization chamber described in the text. The arrow indicates the direction of the long axis of the chamber. The relative response in a given direction is proportional to the radial distance of the curve from the center.

chamber, which had previously been calibrated against our free air standard chamber.

Fig. 2 displays the results of sensitivity measurements as a function of half value layer for conducting polystyrene chamber type *A* and the Baldwin–Farmer chamber type BD11. The shaded points are those obtained with the constant potential generator as a radiation source. The remaining points were obtained with the OEG60 tube and half wave rectification.

The directional response plotted in Fig. 4 was obtained with radiation which had a half value layer of 1.8 mm Al (27.5 keV). The chambers were supported with the long axis horizontal in a V-shaped groove in the upper end of a cylinder of stiff paper. They were positioned so as to be at least 50 cm from any scattering objects. The assembly could be rotated so that the long axis of the chamber made any desired angle with the direction of the beam.

In the diagnostic use of X-rays, exposure dose rates as high as 10 r/sec may be employed in the useful beam although they are usually less than 2 r/sec. The exposure dose rate response for chamber *A* was measured precisely (deviation of 1 per cent detectable) at rates up to 14 r/sec, and less accurately (deviation of 10 per cent detectable) up to 40 r/sec.

Two separate methods of measurement were used. In one, the chamber response was

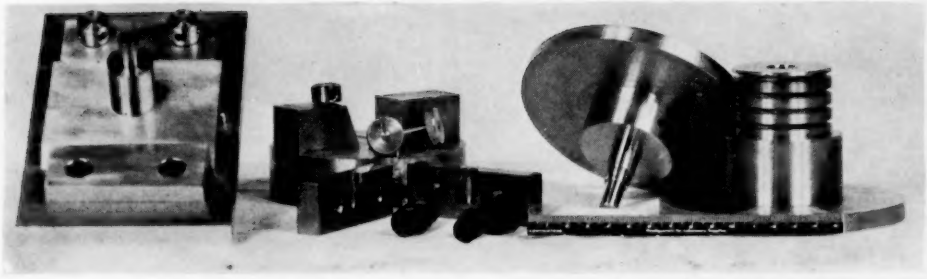

FIG. 5. Photograph of two of the molds being used for quantity production of the type A chamber described in the text. Note that a split mold was necessary for that half of the chamber which has the thread on the outside. Removal from the mold is not otherwise possible. The other half, however, may be screwed off the mold after being formed.

compared with that of a 25 r Victoreen r-meter at dose rates varying from 0.6 r/sec to 14.0 r/sec. The 25 r Victoreen r-meter response is essentially independent of dose rate up to 50 r/sec.[6] In this comparison the tube potential and geometry were kept constant and the variation of dose rate was obtained by changing the tube current. The total dose per exposure was kept constant and at a value such that for all dose rates the chamber potential was reduced to one-half of its initial voltage (250 V). This is necessary since at the highest dose rates used the average collection efficiency may vary when the final voltage is less than one-third of the initial voltage. These measurements showed the response to be constant within a standard deviation of $2\frac{1}{2}$ per cent up to 14 r/sec.

The chamber dose rate response was also studied as a function of collection potential. The dose rate was kept constant at about 6 r/sec and the initial chamber potential was varied from 300 V down to 80 V. Here again the exposure times were adjusted so that the chamber potential was reduced to one-half its initial value. Fig. 6 shows the results of these measurements (it was not possible to make reliable measurements with initial potentials of less than 80 V). These results indicated that for this particular chamber the mean sensitivity was 0.397 V/mr with a standard deviation of ± 1 per cent. This agrees with the values obtained for fixed initial chamber voltage both for the absolute magnitude of the response as well as for the variation of individual readings.

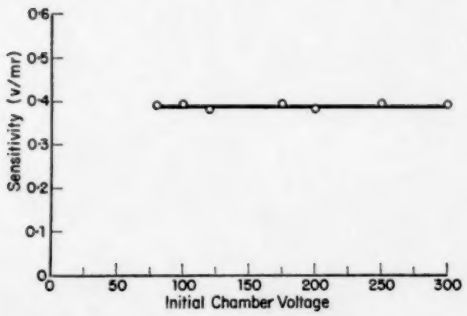

FIG. 6. Sensitivity of chamber A as a function of the initial voltage for a constant dose rate of 6 r/sec. For all measurements the exposure time was adjusted so that the chamber potential was reduced to one half its initial value.

BOAG[3] has treated the problem of collection efficiency for plane and cylindrical chamber geometries and for the extreme conditions of constant intensity and instantaneous pulse radiation. Our case in which the pulses are approximately sinusoidal and are at the rate of 120/sec should be more closely approximated by his continuous intensity case. This is because the pulse duration is large compared with the drift time of the ions across the chamber. Substitution of our chamber characteristics into BOAG's theoretical results for constant intensity yields a collection efficiency of not less than 0.99 for dose rates up to 40 r/sec. For instantaneous pulses this efficiency would be maintained up to dose rates a little above 4 r/sec. Our experimental results are not inconsistent with the calculated values for these conditions.

MANUFACTURE

All the chambers described above have been manufactured by machining solid conducting polystyrene stock. This method is expensive, time consuming, wasteful of material and the dimensions may be reproduced only to within an accuracy of $\pm 1/1000$ in. Accordingly a molding method has been developed and other chambers are presently being made by this technique (see Fig. 5). The authors wish to acknowledge gratefully the assistance of Dr. FAILLA, who advised them of the commercial availability of conducting polystyrene, and of Dr. ROSSI who advised on molding apparatus.

Four separate molds are required: one for each end, one for the collecting electrode and one for the insulator. In all cases the mold is mounted in a press and heated to just above the flow temperature of 250°F. Conducting polystyrene chips are weighed out and added to the female mold. A pressure of not more than 30 lb/in^2 is sufficient. After cooling to 195°F the chamber section may be removed from the mold and is ready for use.

CONCLUSION

A miniature ionization chamber has been developed which satisfies the various critical

criteria necessary for exposure dose measurement in the diagnostic X-ray region. Chambers with completely reproducible properties can be made in large quantities by a molding technique.

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INTEGRITY OF VINYL PLASTIC SUITS IN TRITIUM ATMOSPHERES*†

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Abstract—Significant amounts of tritium oxide penetrate vinyl plastic suits when exposed to high level concentrations. The degree of penetration can be estimated from data obtained during control tests. Exposures of personnel using the suits can be controlled by establishing working time limits based on the expected penetration. Laundering does not appear to affect the overall permeability of the plastic. The test data permit extensive use of the inexpensive two-piece suit, resulting in considerable cost savings in a tritium protection program.

INTRODUCTION

TRITIUM oxide is produced in heavy-water-moderated reactors as a result of neutron capture by deuterium. The oxide, the most hazardous form of the isotope, unlike most radioactive materials, is assimilated by absorption through the skin as well as through the respiratory system.

Although data collected by PINSON and LANGHAM indicate that assimilation through the skin may be less than that through the respiratory system, they conclude that both modes of entry into the body are of approximately equal importance.[1] Protection of personnel working in contaminated atmospheres must necessarily include consideration of both modes of entry. The problem of personnel protection is further complicated since tritium oxide, acting similarly to water vapor, penetrates to some degree many plastic and rubber materials commonly used in the fabrication of protective equipment.

Work in low-level tritium-contaminated atmospheres is permitted without respiratory or body protection, providing time limits are established within which body assimilation will not exceed the maximum permissible amount of tritium allowable in the body. (A reasonable operating limit to assume is 1 mc in the entire body.) Time limits are calculated for a given atmosphere by estimating assimilation through the respiratory system, based on 100 per cent absorption, and assuming equal absorption through the skin. This method is convenient and provides a limited safety factor. Frequently, working time limits are doubled by the addition of respiratory protection. When work is to be performed in tritium concentrations of about 1000×10^{-5} $\mu c/cm^3$ of air, the time limitation is too short for practical purposes (calculations show that 1 mc would be absorbed in 2.4 min) and additional protection must be used.

DISCUSSION

Protective suits

At the Savannah River Plant, one-piece (12 mils thick) and two-piece (6 mils thick) polyvinyl chloride plastic suits are used for protection. These suits, shown in Figs. 1 and 2, are made by the Snyder Manufacturing Co., New Philadelphia, Ohio.

The plastic provides a barrier between the tritium atmosphere and the body. For breathing purposes, air is continuously supplied to the suits at a rate of from 6 to 8 ft^3/min, and is exhausted from the one-piece suit through five diaphragm exhaust valves. The user enters and exits from the one-piece suit through a double

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zippered opening on the back. The two-piece suit consists of a jacket and pants which are tightened at the waist by drawstrings. Air is exhausted from this suit at the waistline. Vinyl straps on the jacket and pants prevent the garments from separating during use.

Literature relating to the permeation of tritium through plastics is essentially nonexistent; therefore, a test program was established to determine the limitations, if any, when wearing these suits.

METHODS

The direct exposure technique was considered the most practical approach for determining the overall integrity of the plastic suits. A box, 8 ft × 4 ft × 3 ft, was fabricated from 1 in. plywood to contain tritium atmospheres for the tests. Two glove ports and a "Lucite" door and window were included in the construction. Joints in the box were gasketed and caulked to prevent leakage. The outside was covered with Amercoat acid-resistant paint, and the inside was coated with a water-base strippable coating (Monsanto D-1000) in order to facilitate decontamination. Electrical outlets, an air supply line, and sampling lines were sealed through the box wall. To prevent pressurizing the box as a result of air exhausting from the suit, a $1\frac{1}{2}$ in. vent pipe was installed to allow air to pass freely from the box to a nearby exhaust hood.

The plastic suits were placed in the box and inflated by continuously supplying breathing air at the normal rate of from 6 to 8 ft^3/min. A supply of tritiated water (containing 1 c tritium/cm^3), a hot plate, and a circulating fan were placed inside the box to produce the desired tritium atmosphere. (The use of tritiated water as the activity source insured that tritium would be in the oxide form.) After the box was sealed, the desired tritium atmosphere was generated by evaporating the water inside the box. The equipment was operated through the glove ports. A rheostat was used to control the temperature of the hot plate and, in turn, the evaporation rate of the water.

A typical curve is shown in Fig. 3. Curves summarizing the data from the three series of tests are shown in Fig. 4.

The tests show tritium oxide permeates vinyl plastic suits. The time lags noted offer a

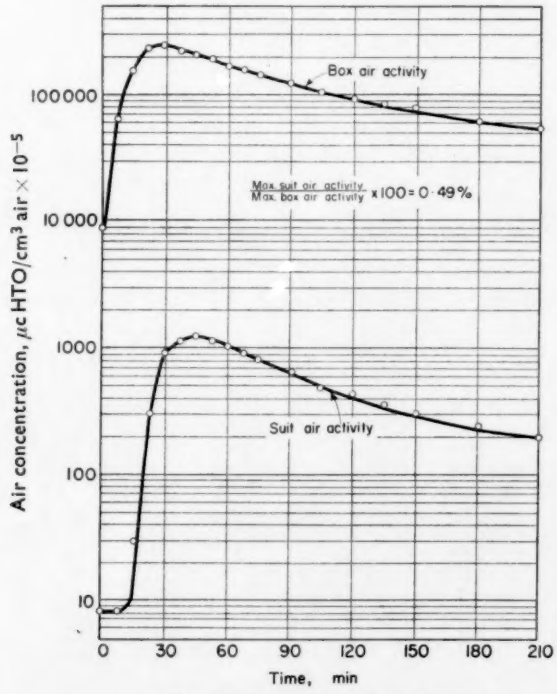

FIG. 3. Typical plastic suit curves.

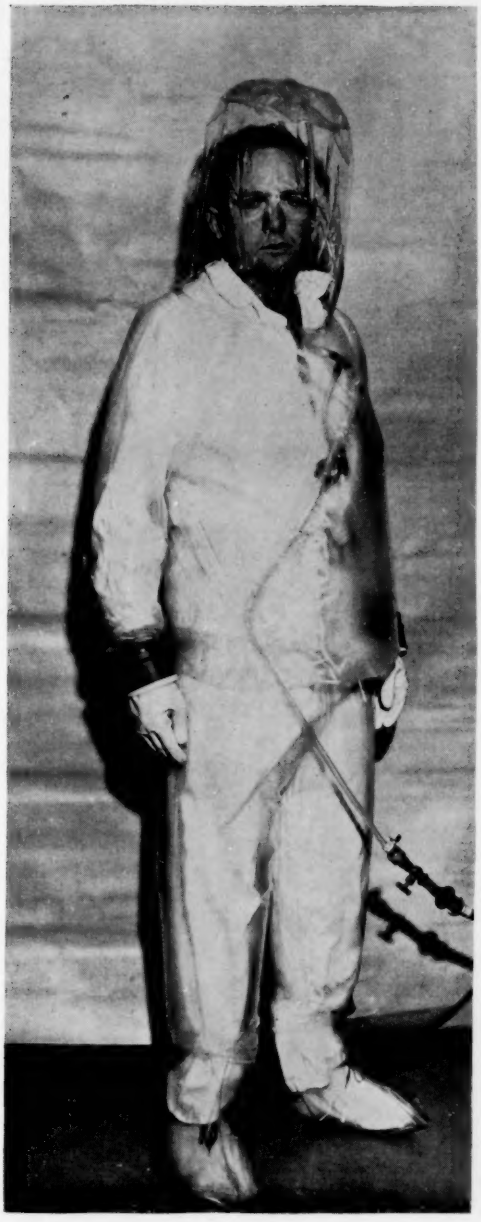

FIG. 1. One-piece suit.

FIG. 2. Two-piece suit.

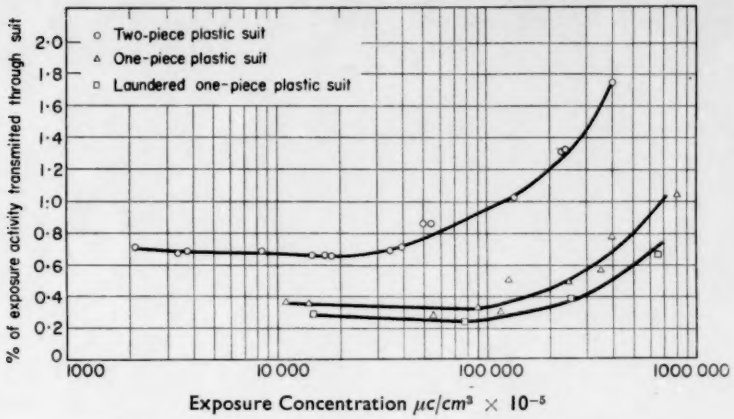

FIG. 4. Tritium oxide permeation through air-supplied suits.

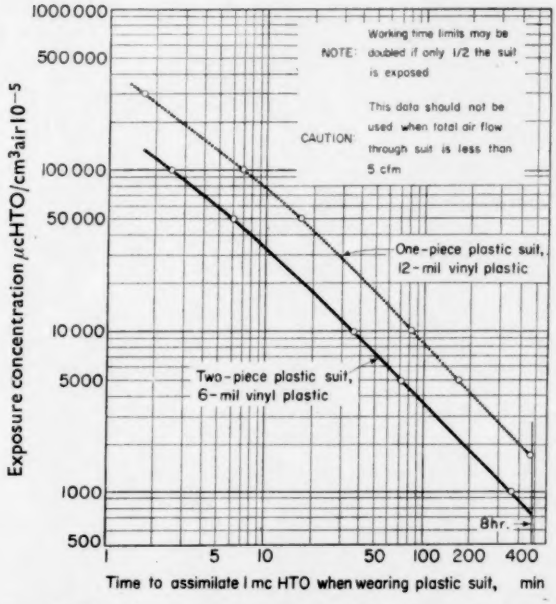

FIG. 5. Time limits for plastic suit use in tritium oxide atmospheres.

protective safety factor if the suits are exposed to sudden bursts of activity. Since the suit was subjected to extreme conditions, i.e. the entire suit was exposed, less penetration would be expected where only parts of the suit are exposed. Laundering of the suit did not appear to affect the overall integrity of the plastic.

To provide a guide for controlling the exposures of personnel, data from Fig. 4 were transposed to a graph (Fig. 5) which shows the estimated time in which 1 mc of tritium would be assimilated when wearing either suit in a specific concentration. The graph serves as a guide to determine which suit should be worn

and the working time limit for that suit. When only one-half of the suit is exposed, the time limit may be doubled.

Data from these tests have provided an effective method of controlling personnel exposures. In addition, extensive use of the less expensive two-piece suit (cheaper by a factor of 7) was permitted which resulted in a considerable cost reduction in the tritium personnel-protection program.

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CHARACTERISTIC CURVES FROM DIFFERENT IONIZING RADIATIONS AND THEIR SIGNIFICANCE IN PHOTOGRAPHIC DOSIMETRY

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Abstract—Characteristic curves for six different emulsions have been obtained with nuclear radiations of a wide range of specific ionization. Sources included α-particles, protons, neutrons, and β-, γ- and X-rays. The two most sensitive films (Eastman Type K and Dupont Type 555) exhibited characteristic curves identical in shape for all exposures. Curves for the remaining films were found to be dependent on the type of radiation. When log density was plotted against log exposure, the curves fell into two groups. For particles with specific ionization equal to or greater than that produced by 30 keV X-rays, the slope in the linear region approached 1.0. For high-energy β-rays, γ-rays and electrons, slopes greater than 1.0 were obtained. In most cases identical saturation densities were observed. Data are presented which show that γ-ray characteristic curves can be simulated with β-rays. This technique can considerably simplify exposure procedures, particularly where high-level exposures are involved.

INTRODUCTION

FILM responds to ionizing radiation by the formation of a latent image in the silver halide grains of the emulsion. Upon development these grains are reduced to metallic silver which, in aggregate, is seen as blackening or increased density. The characteristic curve of a film is a systematic representation of the relationship between density and the amount of exposure. Some commonly used radiation sources are radium and Co^{60} γ-rays, high- and low-energy X-rays, and β-rays. Films of widely varying sensitivities have been used for determining exposure from these sources. This report is concerned with the study of characteristic curves produced by the different ionizing radiations. It is further hoped to obtain a better understanding of film response and in this manner strengthen radiation dosimetry techniques. The specific objectives studied were:

(1) Film response to radiations of different specific ionization including neutrons, α-particles and protons.

(2) Dependence of response on emulsion sensitivity.

(3) Influence of developing time on the characteristic curve of a given emulsion.

APPARATUS AND TECHNIQUES

Radiation used included γ-rays, β-rays, electrons, X-rays, neutrons, protons and α-particles. Table 1 enumerates the sources, their energies, and their relative specific ionization. Except for the electron, proton and α-beams the latter values are approximate. For X- and γ-rays, and neutrons they are based on the specific ionization associated with the average energy of the secondary electrons or of the recoil protons which are produced.

Co^{60} exposures were obtained with a source calibrated by the National Bureau of Standards. During exposure each film packet was covered on both sides by a cardboard 160 mg/cm^2 thick which was in turn covered by a lead filter 0.89 mm thick. The filter minimized the effects of low-energy scattered radiations. Secondary electrons originating in the lead were absorbed by the cardboard which is a tissue equivalent material of sufficient thickness to provide electronic equilibrium.[1] The same film holder was also used for neutron exposures.

A 250 kV Westinghouse constant potential therapy unit served as a source of low-energy X-rays. During operation fluctuations were kept to a minimum by

Table 1. *Physical characteristics of the radiation sources used for film exposures*

Radiation source	Type of radiation	Effective energy (MeV)	Approximate* specific ionization
Van de Graaff generator	electrons	1.9	1.0
Thick Sr^{90}–Y^{90} plaque	β-rays	0.7 MeV average	1.2
Co^{60}	γ-rays	1.3	1.4
250 kV constant potential X-ray generator	X-rays	0.180	7
		0.030	12
University of California 184 in. frequency modulated cyclotron	protons	340	1.6
	α-particles	380	18
University of California 60 in. cyclotron	neutrons:		
24 MeV deuterons	Be^9 (d, n) B^{10}	9	50
12 MeV protons	Be^9 (p, n) B^9	2	100

* Minimum specific ionization is designated as 1.0. For neutrons, X- and γ-radiation, the given values are for the secondary ionizing particles.

means of four voltage regulators. Films exposed to X-rays were not shielded.

Beta-ray exposures were made using a multiple Sr^{90}–Y^{90} source unit similar to one described by DUDLEY[2]. By successively diluting solutions of Sr^{90}–Y^{90}, six samples of decreased activity were obtained. Each solution was then mixed with a fixed amount of dental plastic and cast within a recess in a lucite block. A $\frac{1}{32}$ in. bakelite filter attenuated the 0.6 MeV Sr^{90} component and limited exposure primarily to beta rays from Y^{90}. Measurement of the resultant spectrum by means of a scintillation spectrometer established the average β-ray energy as 0.7 MeV. A 2 MeV Van de Graaff generator was used as a source of 1.9 MeV electrons. All electron and β-ray film exposures were made under maximum backscatter conditions.

Neutron exposures were made at the University of California 60 in. cyclotron. Two neutron spectra described in an earlier report were used.[3] Fast neutrons with mean energies of 2 MeV and 9 MeV were produced by bombarding a thick target with 12 MeV protons and 24 MeV deuterons, respectively. Exposures were made 5 in. from the target in order to minimize blackening from stray background radiation.

The 380 MeV α and 340 MeV proton beam exposures were obtained at the University of California 184 in. frequency-modulated cyclotron. Physical properties of the particle beam have been described by TOBIAS *et al*[4].

All films not otherwise described were developed for 5 min with constant agitation at 68°F. Eastman liquid X-ray developer and fixer were used throughout. Densities were read on a Macbeth–Ansco color densitometer, Model 12, which covers a range of densities from zero to six in two steps. Double-coated film may be read up to a density of 12 if the emulsions on each side are read separately and the net densities added.

With the exception of Eastman Translite, only dosimeter-type films were studied. They are Dupont Types 606, 510, 502 and 555; and Eastman Type K. Type 606 and Translite, the two least sensitive emulsions, are similar in response although Translite is a double-coated projection printing emulsion on a translucent base while Type 606 is a single-coated emulsion on a clear base. Type 510 is a film of intermediate sensitivity. Type 502 is a sensitive emulsion designed for use with intensifying screens, while Types 555 and K represent film of maximum sensitivity. These last three emulsions are most useful in monitoring low-level radiations. Table 2 lists the films and the Co^{60} exposure that will produce a net density of 1.0.

A characteristic curve relating density D to exposure E is customarily obtained by plotting D against log E in the form introduced by HURTER and DRIFFIELD. For some purposes a plot of D against E or log D against log E presents the data in more convenient form.[5]

Characteristic curves in this report have been plotted as log D vs. log E. In observing the curves of Figs. 1–7, three distinct regions are noted: the straight line or "linear" region, the shoulder of the

Table 2. Gamma-ray film sensitivity of six photographic emulsions

Film	Co^{60} exposure for net density of 1.0 (r)
Eastman Translite	98.0
Eastman Type K	0.5
Dupont Type 606	90.0
Dupont Type 510	16.0
Dupont Type 502	1.6
Dupont Type 555	0.9

curve and the saturation region corresponding to development of all the available grains. As will be shown later, the slope at low densities may be used to evaluate the relative grain sensitivity for particles of different specific ionization.

Because of the large differences in the dosage sensitivity of film to different ionizing particles, all curves are plotted on a relative basis. Films are some ten to twenty times more sensitive to low-energy X-rays than to γ-rays; for neutrons the sensitivity for certain exposures decreases to 0.01 of their γ-ray response.[6,7,8] Wherever two curves were directly compared they were normalized to the Co^{60} curve at the shoulder region.

EXPERIMENTAL RESULTS

Initial studies were made on the effect of developing time on characteristic curve response. Fig. 1 shows curves obtained with 502 film exposed to β-rays and developed for 3, 5, 7.5 and 10 min at 68°F. As developing time was increased a proportionate increase in emulsion sensitivity was observed in the linear region, whereas the saturation densities were found to be independent of processing conditions. From the relationship of the curves it is seen that beyond a given developing time there is no further increase in effective film sensitivity. Thus, except for changes produced in the shoulder region, the shape of the characteristic curve will not be influenced by nominal differences in developing conditions.

The effect of different specific ionizations becomes apparent when characteristic curves are drawn for low-energy X-ray and Co^{60} γ-ray exposures. Fig. 2 is such a curve for Type 606 film. Below a net density of two, two distinct slopes are observed. The slope for Co^{60} is steeper than for 30 keV X-rays. Above this density the curves have the same shape and very nearly the same saturation density. Fig. 3 shows a similar set of curves for Translite. The same relationship between the

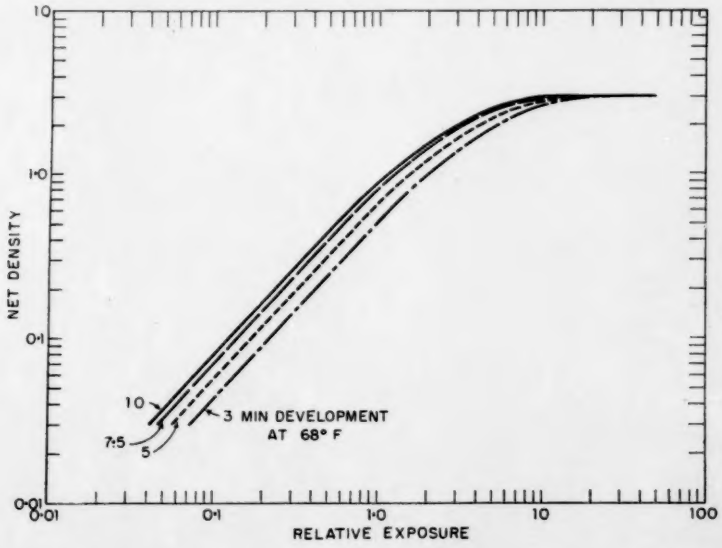

FIG. 1. Effect of 3, 5, 7.5 and 10 min development on Type 502 film.

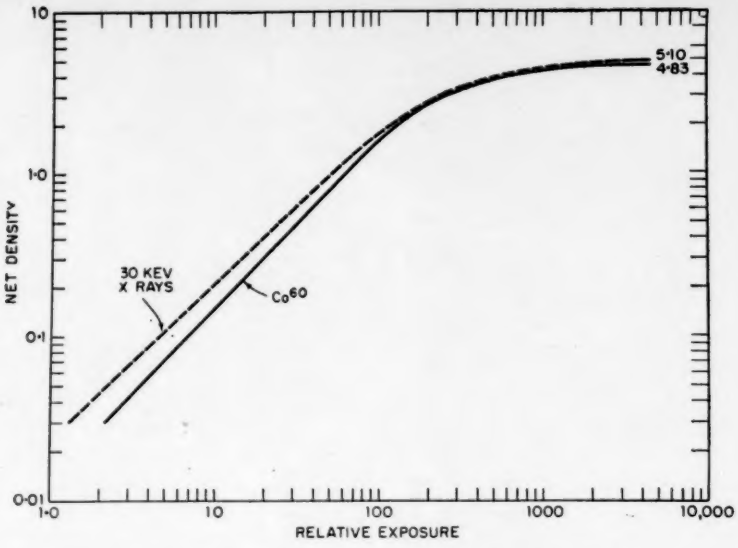

FIG. 2. X- and γ-ray characteristic curves for Type 606 film.

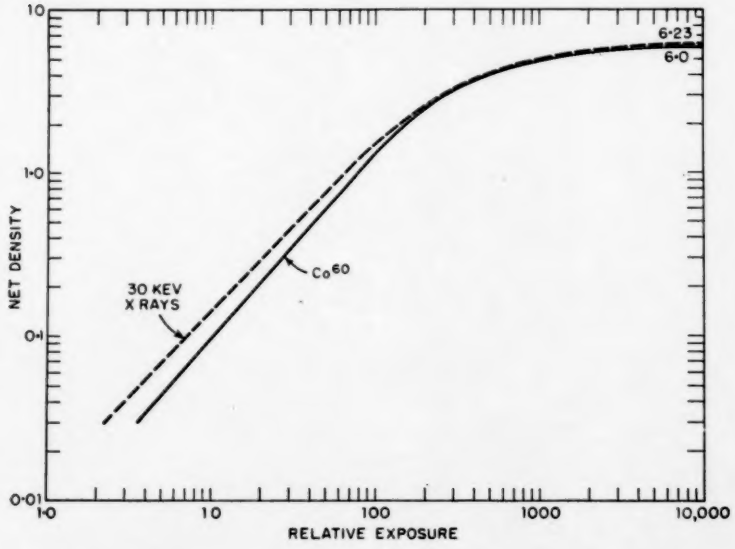

FIG. 3. X- and γ-ray characteristic curves for Translite film.

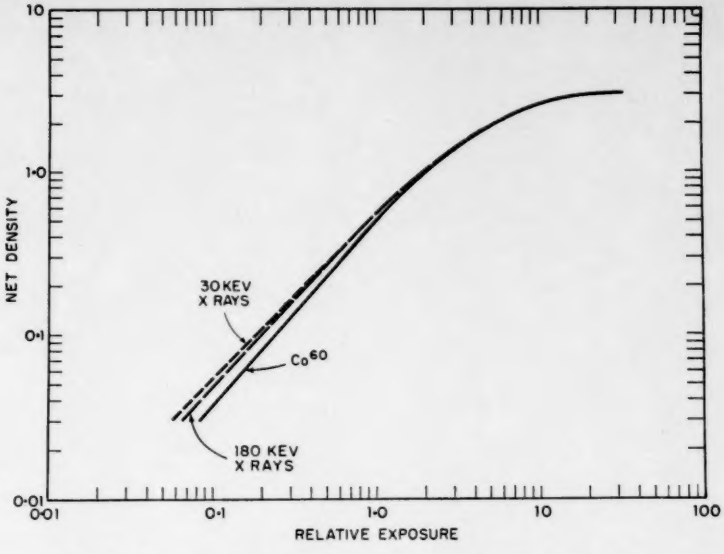

FIG. 4. X- and γ-ray characteristic curves for Type 502 film.

X-ray and Co^{60} points is seen to exist. In Fig. 4 characteristic curves are plotted for Type 502 film exposed to 30 and 180 keV X-rays and to Co^{60}. The additional plot illustrates the response at an intermediate value of specific ionization. Although 502 is a considerably more sensitive emulsion than either 606 or Translite, the response characteristics of all three emulsions are notably similar. In every case a greater slope was obtained for the less heavily ionizing radiations.

Neutron characteristic curves obtained for the three films were identical in shape with those for 30 keV X-rays as shown in Figs. 2, 3 and 4. No differences were detected between the two neutron energies at which exposures were made (Table 1).

Exposures made with Sr^{90} β-rays and 1.9 MeV electrons produced curves similar to that of high-energy γ-rays. Fig. 5 is a curve for 502 film in which experimental points have been plotted for Sr^{90} and Co^{60}. A single curve may be fitted to all points plotted over a net density range from 0.04 to saturation density at 3.0, corresponding to γ-ray exposure values from 0.1 to 50 r. Points obtained with 1.9 MeV electrons were also found to lie on the curve. A similar agreement between Sr^{90} and Co^{60} points for Translite is shown in Fig. 6. An X-ray curve for Translite film is also plotted and points from 340 MeV proton and 380 MeV α-particle exposures are shown normalized at the shoulder of the X-ray curve. The alpha data follow the curve while the proton points are seen to diverge at the lower densities. Although relatively few experimental points were obtained with the respective particle beams, the data are consistent with the pattern previously established in that the less heavily ionizing protons produce steeper slopes. The two curves of Fig. 6 are not directly comparable since each group of films was of a different emulsion batch and was processed at a different time.

In comparing the effects of the different radiations on photographic emulsions, two trends become evident. The slopes of the curves attain maximum values for particles approaching minimum specific ionization, i.e. electrons, β-rays and γ-rays, and become less steep at greater specific ionization. It is further observed that the slope is essentially unity for 30 keV X-rays. Beyond this point the characteristic curve maintains its identity independent of any further increase in specific ionization.

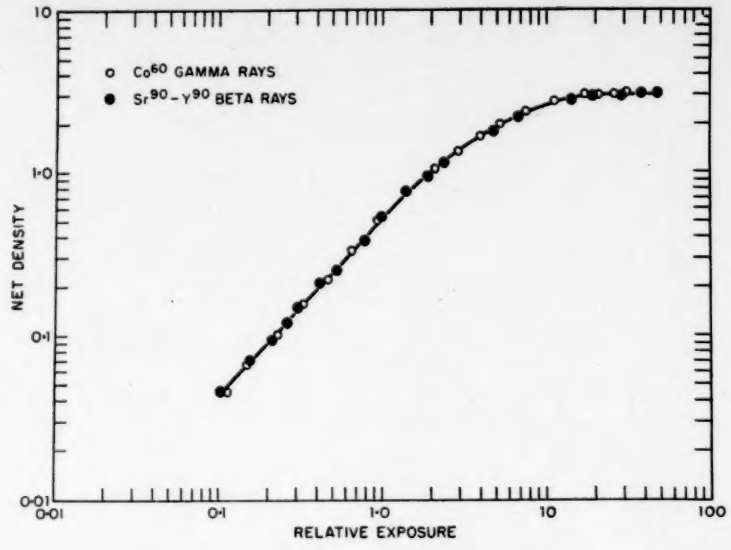

FIG. 5. Experimental points for Type 502 film.

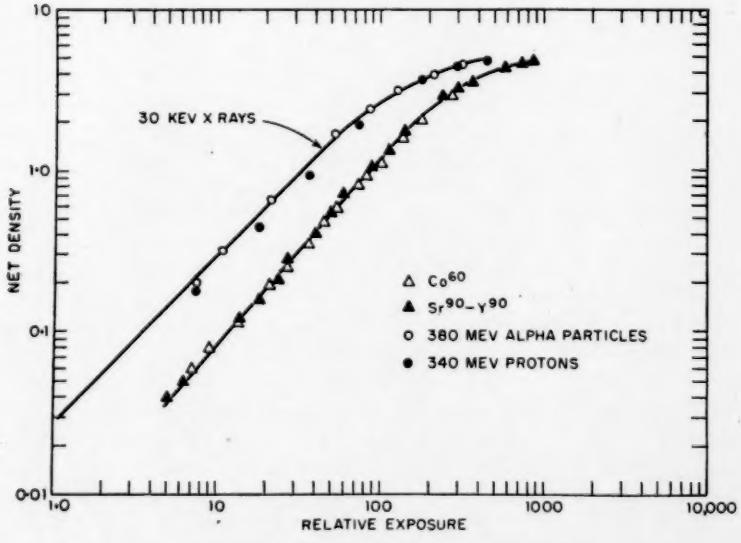

FIG. 6. Experimental points for Translite film.

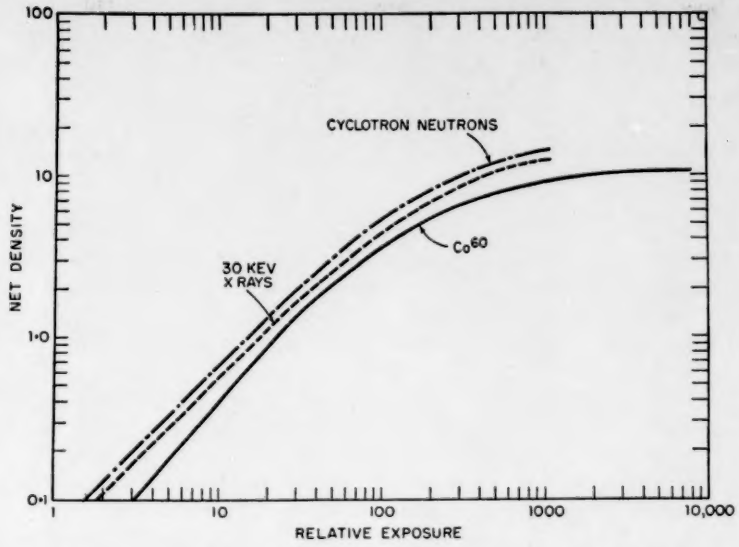

FIG. 7. Neutron, X-ray and γ-ray characteristic curves for Type 510 film.

Types K and 555, the two most sensitive films, exhibited results different from those previously described. In this case identical characteristic curves were obtained for all exposures including high-energy γ-rays and electrons. The slope in the linear region was 1.0 for both films.

Characteristic curves for 510 film obtained with Co^{60}, low-energy X-rays and neutrons, exhibited unusual results in that saturation density was found to be strongly dependent on specific ionization. The three curves are compared in Fig. 7. A saturation density of eleven was obtained for γ-rays. Saturation densities for X-rays and neutrons occurred beyond the limits of the densitometer and for this reason could not be determined. For both curves an approach to saturation was indicated, with the neutron curve attaining higher values. In the low density region both neutrons and X-rays showed parallel slopes of approximately 1.0. The slope was greater for γ-rays, β-rays and electrons, all of which gave identical curves.

Subsequent checks on the influence of developing time on saturation density revealed that complete development was not attained under the processing conditions that were used. Unlike the data of Fig. 1 for 502 film, saturation densities for Co^{60} exposures were found to increase progressively with 3, 5, 7.5 and 10 min development. Maximum densities ranged from 8.1 at 3 min to values greater than 12 at 7.5 and 10 min.

The results indicate that incomplete development was largely responsible for the unusual behavior of 510 film. This implies that either the processing was stopped before all the grains were developed or that the developed grains were not allowed to attain maximum size. An additional series of neutron exposures suggested that in this case full development was reached with 7.5 and 10 min developing times, whereas this clearly was not so for the less heavily ionizing γ-rays.

DISCUSSION

The response of film to monokinetic β-particles from 0.033 to 1.8 MeV has been investigated by DUDLEY,[2] who concluded that the resultant characteristic curves were independent of energy. Experimental data were plotted by DUDLEY in the same manner as in the present report. He observed that density–exposure relationships in the linear portion of the curve could be described by the equation $D = kE^n$, where k is a constant and n is the

slope. The net density essentially shows how many grains are made developable for a given exposure. If all grains are of the same size, then the value of n is defined as the number of hits per grain required for development. Since photographic emulsions exhibit wide variations in grain size, this idealized condition cannot be achieved.

TOCHILIN *et al.*[8] have studied the dosage sensitivity of dosimeter films over a wide range of specific ionization. From the observed similarity between dosimeter films and electron-sensitive nuclear emulsions it was concluded that any singly-charged particle passing through an average grain will impart sufficient energy to render it developable.

Film sensitivity to ionizing particles is uniquely determined by the amount of energy expended in the grain; consequently, film sensitivity is related to grain size. The larger grains within the emulsion become developable at lower exposures and affect the shape of the curve at the lower densities.

Types K and 555 film are examples of large-grain, sensitive X-ray emulsions. Unit slope for these films indicates that all the grains along the path of a particle become developable regardless of specific ionization.

Slopes greater than 1.0, obtained for the remaining films with β- and γ-ray exposures, indicate that particles of minimum specific ionization will not impart sufficient energy to render an average grain developable in every case. The slope decreases progressively for particles of greater specific ionization and approaches a value of 1.0. When used as photographic dosimeters these films require a calibration source comparable in specific ionization with the radiation to be detected, if ultimate accuracy is to be obtained. For this reason Type K and 555 films, whose response is independent of energy, are better suited for use as dosimeters in mixed radiation fields.

One practical application resulting from this study is that characteristic curves for γ-rays can be obtained with β-ray plaques calibrated with film to indicate an equivalent γ-ray exposure. A group of sources with several levels of activity will allow a complete curve to be reproduced in a short period of time. The required activity is low and sources equivalent to large sources of γ-rays can be used directly in the laboratory without need for elaborate shielding.

CONCLUSIONS

(1) Characteristic curves of identical shape were obtained for all films with β-rays, γ-rays and electrons. For this reason characteristic curves for γ-rays may be obtained with β-sources calibrated in terms of equivalent γ-ray exposure.

(2) For Types 502, 606 and Translite film the shape of the characteristic curve in the low density region was found to be dependent upon specific ionization. It is emphasized that all curves are normalized on a relative basis rather than on absolute dosage sensitivity.

(3) For Types K and 555 film the shape of each characteristic curve was found to be independent of specific ionization, therefore these films are better suited for use as dosimeters in mixed radiation fields.

(4) For all films, excluding Type 510, it was concluded that complete development was obtained under standard processing conditions with X-ray developer. Nominal changes in the developing conditions did not greatly influence the shape of the characteristic curves.

(5) Changes observed in the saturation density of 510 film with different exposure conditions and developing times indicated that complete development was not achieved.

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A FILM TECHNIQUE FOR MEASURING THE EXPOSURE DOSE FROM PLUTONIUM*

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Abstract—The electromagnetic radiations from plutonium are considered in three effective energy groups. The first group consists of X-rays with an effective energy of about 17 keV. The second group consists of γ-rays with an effective energy of about 60 keV. The third group consists of all γ-rays with energies greater than 400 keV. The low penetrating power of the group (I) X-rays is demonstrated, using a water phantom. Because of the ease with which they are absorbed, the testes are considered the critical organ. The measurement of all three energy groups with monitoring film is complicated by the energy dependence of the photographic emulsion used. The energy dependence of DuPont Type 502 emulsion is shown to peak in sensitivity at about 45 keV. The effect of shielding the film, as in a film badge, improves the energy dependence but limits the minimum energy which can be measured to about 60 keV.

The shielding features in the Hanford Film Badge permit measurement of the exposure dose of the three energy groups. From a set of generalized equations relating exposure dose and net density, expressions are derived for the dose due to each energy group.

THE dose from plutonium is primarily due to α-particles emitted from material inside the body during radioactive decay. Since α-particles are easily absorbed, a general misconception prevails that the dose from plutonium is negligible when these particles are contained by minimum shielding. As a consequence, most radiation protection measures are directed toward eliminating the α-particle problem with little or no regard for the exposure dose arising from other radiations. Radiations associated with reactor-produced plutonium include neutrons, β-rays, X- and γ-radiations.

At Hanford, the neutron dose is measured by use of nuclear track emulsions[1] in a manner similar to the one described by Cheka. This technique is primarily for neutrons with energies greater than about 0.7 MeV. Further work on personnel monitoring for lower energy neutrons is in progress. Beta-rays are not a major problem in whole body dosimetry since, during normal operations, all plutonium work is accomplished with adequate shielding against this type of radiation. It is the purpose of this paper to describe the measures developed by many individuals over a number of years, in evaluating the contribution to the exposure dose of the X- and γ-radiations from plutonium.

Chemically pure plutonium is seldom encountered. In general, it is contaminated with daughter and fission products to varying degrees. For daughter products, the degree of contamination depends on the fractional amounts of the various plutonium isotopes initially present and on time since chemical separation. The isotopic composition depends upon the length of reactor irradiation time[2] as shown in Fig. 1. For example, a reactor exposure of 6000 MW-days/ton of uranium results in a plutonium composition of 65 per cent by weight of Pu^{239}, 25 per cent of Pu^{240}, and 10 per cent of Pu^{241}. For an exposure of 12,000 MW-days/ton, the percentages are 53 per cent,

* This paper is based on work performed under contract No. W–31–109. Eng. 52 for the Atomic Energy Commission.

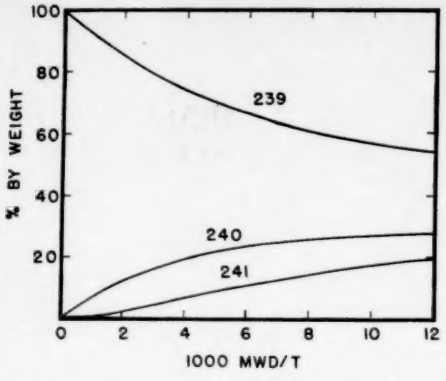

FIG. 1. Isotopic composition of plutonium as a function of reactor exposure per ton of uranium.

27 per cent, and 20 per cent, respectively. Since each of these isotopes emit radiation of different energies, it follows that the dose rate from reactor-produced plutonium depends in part upon the reactor exposure.

The surface absorbed dose rates from the isotopes have been estimated by ROESCH[3] and are summarized in Table 1. The energies which have been identified are noted here also. The contribution from Pu^{241} becomes more significant as the time since separation increases. These calculations do not take into account the dose contribution from fission product contaminates remaining after chemical separation. However, Fig. 2 shows the variation of the surface dose rate with reactor exposure at 5 days after chemical separation. More than 70 per cent of the total dose is due to X-rays with energies less than 20 KeV. None of these calculations take into account shielding.

The shielding afforded by production hoods at Hanford is a nominal $\frac{1}{2}$ in. thick slab of lucite. From absorption measurements through this shielding, the energies from reactor-produced plutonium fall into three effective energy groups. The first of these is a group of X-rays with an effective energy of about 17 keV. The second and third groups consist of γ-radiation with effective energies of about 60 keV and 400 keV, respectively. The relative exposures contributed by each of these group changes with reactor exposure, time since separation of the plutonium, and the geometrical form of the plutonium. However, the effective

Table 1. *X- and* γ*-radiations from* Pu *isotopes*

Isotope	Type radiation	Energies (MeV)	Surface dose (rads/hr)
Pu^{238}	γ	0.04	2
	X-rays	Unidentified	980
Pu^{239}	γ	0.039 0.053 0.100 0.124 0.384	0.056
	X-rays	0.0136 0.0174 0.0205	0.61
Pu^{240}	γ	0.04	0.37
	γ (fission product from spontaneous fission)	~1.0	0.25
Pu^{241}	X-rays	Unidentified	14
β / α Am^{241}(>99%)	γ	0.026 0.043 0.060 0.099	$220(4.28 \times 10^{-4}t)$*
	X-rays	0.017	240
U^{237}(≪1%)	γ	0.059 0.207 0.334	$23(1 - e^{-0.102t})$*

* For $t \ll 5000$ days.

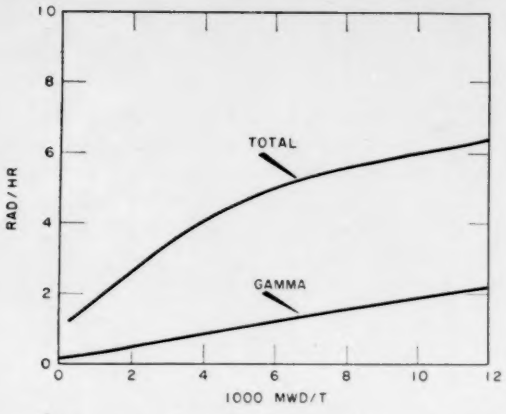

FIG. 2. Surface dose rate of plutonium at 5 days after chemical separation as a function of reactor exposure per ton of uranium.

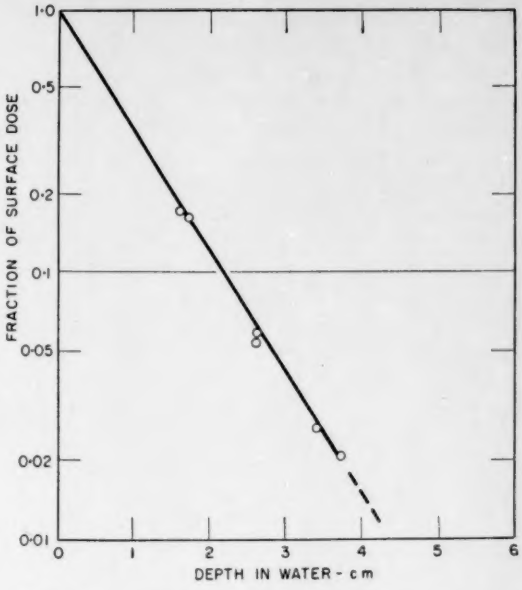

FIG. 3. Absorption of 16.1 keV X-rays in a water phantom.

energies, especially of the first two groups, change very little.

Before discussing film measurements, it is of interest to discuss the absorption of the low energy X-rays comprising group (I). These X-rays are found to be easily absorbed by materials with large effective atomic numbers; but numerical data concerning their absorption were not complete, especially in tissue-like material. Therefore, water phantom measurements were undertaken.

The source of X-rays used for this low energy of about 17 keV was a K-fluorescent X-ray source designed by LARSON *et al.*[4] This source is capable of producing very nearly monoenergetic X-rays when used with a suitable target. In this case, a zirconium target was used to give 16.1 keV in energy. The detectors used were hermetically sealed pocket ionization chambers (Victoreen).

The data shown in Fig. 3 indicate that the absorption of these X-rays corresponds to a 50 per cent reduction at about 0.8 cm of water, and less than 10 per cent remain at a depth of 2.3 cm. On the basis of these measurements, the dose at the depth of the blood forming organs is estimated at less than 1 per cent of the surface dose. The dose at the depth of the gonads (1 cm) is estimated at 35 per cent of the surface dose and at the depth of lens of the eye, 73 per cent of the surface dose. Therefore, if protection is provided the eyes, the gonadal dose is limiting.

Safety goggles made of glass were found to afford adequate eye protection for the group (I) X-rays. Therefore, if one now considers the testes as the critical organ, the contribution of these X-rays, is reduced to about 10 per cent of the combined surface dose at a depth of 1 cm in tissue.

Early attempts at film measurement of the low energy group consisted of using thin gold foils and silver shields. These measurements were partly successful in that it was possible to discriminate between β-rays and soft X-rays. This general principle has been incorporated in the recently developed Hanford film badge[5] and permits measurement of all three energy groups. This badge contains three shields and an unshielded portion called the "open window". Two of the shields are designed to be of equal mass per unit area but different Z, viz. 0.13 g/cm^2 of aluminum and silver. The third shield is a nominal 1 g/cm^2 thickness of silver.

The spectral response of DuPont Type 502 emulsion as measured by LARSON[6] is shown in

Fig. 4. From this curve, the ratio of true dose to that obtained from density measurements behind the thick silver shield is very nearly 1.0 for energies above 200 keV. The decrease in the ratio below 200 keV is in semiquantitative agreement with that expected from an ion chamber filled with AgBr and emulsion molecules but with walls too thick to satisfy the Bragg–Gray conditions.[7] This ratio continues to decrease reaching a minimum at about 130 keV behind the thick silver shield and at about 45 keV without external shielding. The increase of this ratio at still lower energies is due partly to the absorption in the silver shield and paper film wrapper, and partly to the onset of the photoelectric effect in air.

For any given energy and with any type of shield, the density produced in the film emulsion is a function of the dose. In general, this function is independent of the energy when suitably normalized.[6] However, the complexity of the film response over the lower energies makes the general problem extremely difficult. The approach taken by KOCHER,[8] makes use of the linear response of DuPont film at all energies. This linearity is good for net densities up to about 0.7. In this case, the net density behind a given shield for photons of discrete energies can be represented by the following equations:

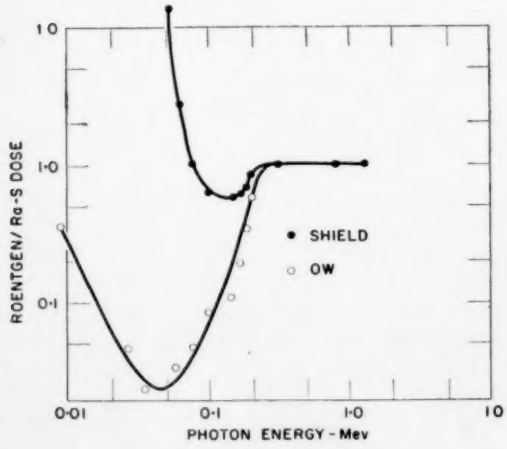

FIG. 4. The energy dependence of DuPont type 502 film.

$$\delta_A = \sum_{n=1}^{K} a_n D_n$$

where

δ_A is the net density behind shield A;
a_n is the slope of the density-dose curve behind shield A;
D_n is the dose due to energy E_n;
K is the number of discrete energies involved.

For shield B, the equations are:

$$\delta_B = \sum_{n=1}^{K} b_n D_n$$

Now, if the number of shields available are equal to or greater than the number of energies involved, the above sets of equations have a unique solution. In this instance, the equations become:

$$\delta_{OW} = a_1 D_1 + a_2 D_2 + a_3 D_3$$
$$\delta_{Ag+} = b_1 D_1 + b_2 D_2 + b_3 D_3$$
$$\delta_{Ag} = c_1 D_1 + c_2 D_2 + c_3 D_3$$

where

OW = "open window" shield;
Ag+ = thin silver shield;
Ag = thick silver shield;
D_1 = dose, 17 keV group;
D_2 = dose, 60 keV group;
D_3 = dose, 400 keV group;

Coefficients a_n, b_n and c_n are the slopes of density–dose curves for a given energy behind shields OW, Ag+ and Ag, respectively, i.e. b_2 = slope for energy group (II) behind the thin silver shield.

It should be noticed that these equations force the density–dose curves to pass through the origin. Measurements indicate that for individual curves, they do not always do so. However, this is considered a measurement error, since on the average the intercept is very nearly zero. Further assumptions are:

$$(1) \quad a_3 = b_3 = c_3 = C$$

$$(2) \quad b_1 = c_1 = c_2 = 0$$

The first assumption states that the density produced by the group (III) energies (400 keV)

is the same behind all shields. The most serious error involved here is the open window density. Poor geometry and medium energies will cause this density to be high by as much as 40 per cent in some cases. However, this results in a conservative answer since the dose due to high energy components is based on the thick silver shield and any excess density produced on the open window is assumed due to the lower energy components.

Assumption (2) states that the X-rays produce zero density behind either silver shield and the 60 keV component produces zero density behind the thick silver shield. This last assumption is very nearly true since it is only when large exposures to the calibration energy for the 60 keV component are made, that measurable densities are found behind the thick silver shield. These densities are so small that they could very well have been produced by scattered radiation from the fluorescent X-ray source. With these limitations in mind, substitution of these assumptions into our equations yields:

$$\delta_{OW} = a_1 D_1 + a_2 D_2 + C D_3$$

$$\delta_{Ag+} = \quad + b_2 D_2 + C D_3$$

$$\delta_{Ag} = \quad + C D_3$$

The solutions for which are:

$$D_1 = \frac{1}{a_1}\left[\delta_{OW} - \delta_{Ag} - \frac{a_2}{b_2}(\delta_{Ag+} - \delta_{Ag})\right]$$

$$D_2 = \frac{1}{b_2}(\delta_{Ag+} - \delta_{Ag})$$

$$D_3 = \frac{1}{C}\delta_{Ag}$$

The parameters a, b and c are found to be quite variable since they depend upon process variables. The ratio a_2/b_2, however, is very nearly a constant equal to 2.1. To obtain the values of a, b and c, a set of calibrated films are developed with each group of personnel films exposed to plutonium radiations. For the group (I) energies, 16.1 keV fluorescent X-rays are used. For group (II), the calibrating energy is 58 keV and for group (III), radium γ-rays are used. In each case, eight exposure doses are used and the parameters determined by fitting the best line through the net density values using the method of least squares.

This method has proved reliable, as has been demonstrated by evaluating a number of audit films exposed to different percentages of the three energy groups. These films were evaluated by routine methods and good agreement was found between the known exposure and the evaluation, even though in some cases the condition of linearity was not met for the low energy groups. For all cases, in which the net density due to all three energy groups did not exceed 1.0, the evaluation was within 15 per cent of the known exposure.

SUMMARY

The electromagnetic radiations from plutonium may be considered in three effective energy groups. Group (I), with an effective energy of about 17 keV consists of X-rays with low penetrating ability. The gonadal dose is limiting for this group with the dose to the critical organ being about 10 per cent of the total dose of plutonium as measured by the film badge. Group (II), with an effective energy of 60 keV consists of a number of γ-rays from various isotopes of plutonium and contaminates. Group (III) has an effective energy of about 400 keV and the problems of film dosimetry for this group are not peculiar to plutonium. A method of measuring the dose for all three energy groups depends upon the measured response of film density with dose. The method is considered adequate for routine use in measuring exposure doses up to 1.0 r.

Acknowledgments—The writer expresses his gratitude to the many people who have contributed to the work presented here. Some of these individuals are mentioned in the references but to single out others would risk omitting a few. However, the writer wishes expressly to thank Mr. J. W. HEALY and Dr. W. C. ROESCH for their constructive criticisms and suggestions in the preparation of this paper.

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NOTE

A Fission Gas Monitor for Gas Cooled or Off-gas Experimental Systems*

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Principle of operation

Induced gaseous activities in a stream of air passing through a high flux zone, namely A^{41} and N^{16}, decay into stable daughter products. Several of the fission gas isotopes produce radioactive daughters. The monitoring system, diagrammed schematically in Fig. 1, utilizes these different decay characteristics of air and fresh fission product gases to determine the presence of fission products in a coolant air stream.

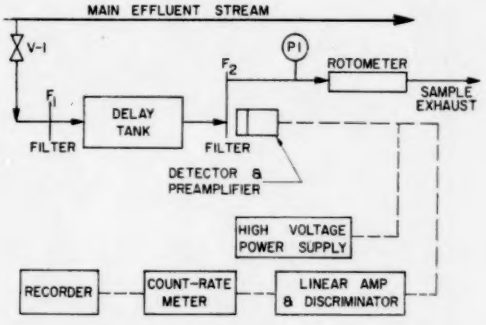


FIG. 1.

The main effluent stream is sampled with a measured flow that is controlled with the valve, V_1. The amount of air flowing through the sample system is determined with the rotometer and pressure indicator. Any system having wide temperature fluctuations would need to have some means of regulating temperature in the by-pass system.

Particulate materials entering the sampler are removed from the air stream by the absolute filter, F_1. All gases enter the delay tank through F_1, remain there for a predetermined time interval and then pass through the filter, F_2, and the rotometer into the exhaust line. The particulate daughters of the radioactive gases that decay while the gases are in the delay tank are collected on F_2. Gamma-radiation from any radioactive material collected on F_2 causes a response of the γ-scintillation detector. This signal, in turn, is amplified and fed to a count-rate meter and recorder. Thus, because of the unique decay characteristics of fresh fission product gases, the presence of fission products in the main effluent air stream is determined by the response of the count-rate meter and recorder of the monitoring system.

Quantitative formulae and sensitivity

The ratio of the rate of entry of a parent gas into the monitoring system to the activity of a daughter product on the filter, F_2, may be expressed mathematically as

$$\frac{A_i f}{D_n} = e^{\lambda i \tau}\left[(\lambda_i+\sigma)(\lambda_i+\sigma-\lambda_n)\right] \div \lambda_i\lambda_n\left[\frac{\lambda_i+\sigma}{\lambda_n}\left(1-e^{-\lambda n t}\right)-\left(1-e^{-(\lambda i+\sigma)t}\right)\right]$$

where

D_n = dis/min of a daughter product (n) on F_2;
A_i = atoms/min of the parent gas (i) produced in the main coolant stream (a constant);
f = the fraction of the main effluent stream entering the monitoring system;
τ = min, delay from production of the parent gas until arrival at filter (F_1);
λ_i = min^{-1}, the decay constant of the parent gas;
λ_n = min^{-1}, the decay constant of the daughter product;
σ = min^{-1}, the reciprocal of the time for a unit volume of gas to flow from F_1 to F_2;
t = min, time interval of sampling after active gas enters the delay tank at F_1.

This formula is based on a constant rate of release of gaseous fission products into the coolant stream and "perfect" diffusion in the sample system, i.e. instantaneous diffusion or mixing of unit volumes of gas and associated daughter products within the delay system. The formula assumes 100% efficiency of the filters, F_1 and F_2, for the collection of the daughter products of the fission product gases and also that "plating-out" of the daughter products on the walls of the delay tank is insignificant. There is some evidence that these assumptions make no

* Work done under contract No. AT(10-1)-205 to the U.S. Atomic Energy Commission, Idaho Operations Office.

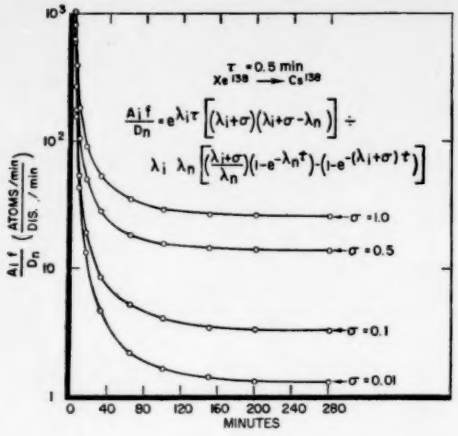


FIG. 2. Ratio of parent entry rate to daughter activity vs. sampling time.

significant error in the calculations at equilibrium. However, these approximations should be checked in each individual monitoring system. The formula breaks down if σ is much smaller than λ_n.

Fig. 2 illustrates the change of the calculated ratio for 17 min Xe^{138} and 33 min Cs^{138} vs. sampling time while using various values of delay time between F_1 and F_2. Because the flow in the delay tank is not "perfect", the accuracy of the graph increases as equilibrium activity of the daughter product is obtained on the filter, F_2. Actually, with a delay time of 10 min between F_1 and F_2, the increase in activity

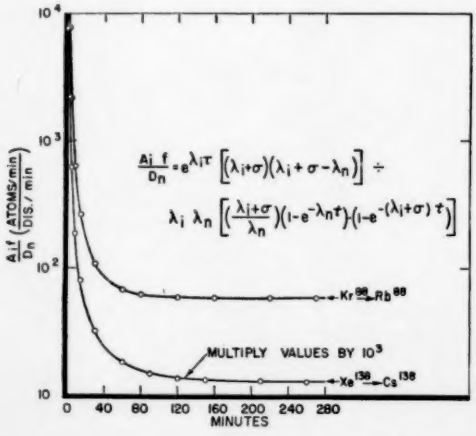


FIG. 3. Ratio of parent entry rate to daughter activity vs. sampling time.
$\tau = 200$ min, $\sigma = 0.1$ min^{-1}.

on F_2 has been observed to rise abruptly in less than 30 sec after the flow of active gas was started through F_1. Notice the very high sensitivity of the system such that the activity of the daughter product on F_2 approaches the rate of production of the parent gas in the sample system.

A monitor utilising a dual detection system, one part for activity on F_1 and the other for activity on F_2, would have the added advantage of a high sensitivity and very quick detection of activities on F_1 while the response of the F_2 detector would be utilized to identify fission products, if present, and to make quantitive estimates of their production rate.

There are numerous combinations of decay chains, delay times to the sampling system, delay times in the monitoring system and methods of analyses of the activity collected on the filter, F_2, which allow variations of monitoring methods to suit the needs of the individual experiment.

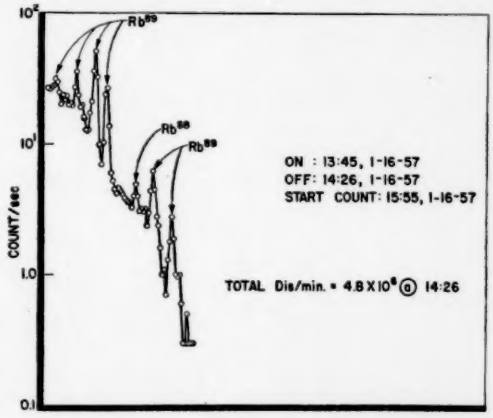


FIG. 4. Activity on F_2 filter downstream of charcoal.

Fig. 3 is an illustration of the selectivity of the instrument for collection of one isotope on F_2. By making τ about 200 min and delay time in the tank of the order of one half-life of the daughter isotope, almost pure Rb^{88} will be collected on F_2. A small contribution from Cs^{138} will be present.

Fig. 4 is a γ-spectra of F_2 activity which was obtained using a charcoal trap to hold up the parent gases before entry into the sample system. At the end of the sampling run the holdup capacity of the charcoal for Xe was not depleted so we see only Rb activities on F_2 from the parent Kr isotopes. The delay times are 24 min in the charcoal and 12 min in the delay tank. The observed spectra is consistent with the holdup time of Kr in the charcoal, the delay time in the tank and complete retention of Xe on the charcoal. Not enough is known about the fractional

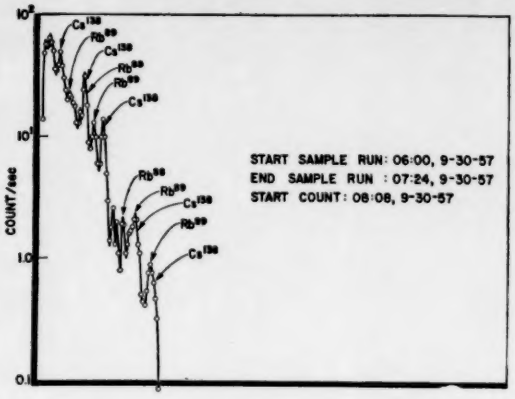


FIG. 5. Activity on F_2 filter downstream of charcoal.

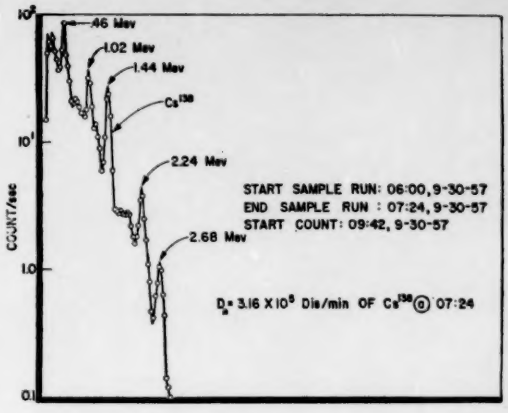


FIG. 6. Activity on F_2 filter downstream of charcoal.

number of γ-counts per disintegration of Rb^{88} or Rb^{89} to determine the disintegration rate of these isotopes from their γ-spectra. The disintegration rate indicated was calculated from a β-decay determination that was started 3 min after flow through F_2 was stopped. The disintegration rate of Rb^{89} could be determined by performing a quantitative analyses of F_2 for Sr^{89} following complete decay of the Rb^{89} on the filter.

Another method for obtaining a single isotope on F_2 for analysis is to keep τ short (about 5 min), and set the delay time between F_1 and F_2 at about 10 min. The activities collected on F_2 in this case will be Rb^{88}, Rb^{89}, Cs^{138} and several shorter life activities. When F_2 is removed for analysis, allow the activity collected to decay for about 2 hr and the resulting γ-spectra obtained will be almost pure Cs^{138}. Fig. 5 is a γ-spectrum of F_2, 44 min after removal from the sampler. Notice the mixture of isotopes still remaining on the filter. Fig. 6 is a spectrum of the same filter obtained 94 min later. The pure Cs^{138} spectrum observed can be analyzed to give the disintegration rate of Cs^{138} on F_2 when the sampling run was stopped.

Recent data obtained with this instrument indicate that when gross fission gases are sampled over periods greater than 2 or 3 hr, it may be necessary to use a small charcoal trap upstream of F_1 to catch iodine and thus prevent the interference of iodine activity with the shorter life rubidium and cesium isotopes on F_2 when quantitative analyses are desired.

SUMMARY

The fission gas monitor is a very sensitive instrument for the detection of fresh fission products in gas cooled or off-gas systems. It is quite versatile so that it can be used on a wide variety of experiments either as a quick, positive and sensitive detector of fission products or as a quantitative measuring device for release of fresh gaseous fission products. Installation costs are reasonable.

The formula presented for quantitative analyses is adequate for constant production rates of fission gases where the delay time between F_1 and F_2 is not much greater than the half-life of the daughter product to be collected and analyzed on the F_2 filter. Filter efficiency and "plating-out" problems should be investigated in each new type of experiment being monitored.

Examples of data obtained with a prototype monitor and curves illustrating calculated sensitivity of the monitor indicate the usefulness of the system for monitoring gas streams containing fresh fission gases and also illustrate a few of the many methods of making quantitative analyses to determine production rates of fission products in gas effluent lines.

Phillips Petroleum Company J. F. SOMMERS
Atomic Energy Division I. J. WELLS
Idaho Falls, Idaho.

Health Physics Pergamon Press 1959. Vol. 2, pp. 216–218. Printed in Northern Ireland

LETTERS TO THE EDITOR

Emergency Monitering Methods for the Determination of the Effective Bone-seeking Fission Products in Milk

A USEFUL categorization of the radioactive products of nuclear fission is by functional group based upon the "critical organ" concept. Thus we have bone-seeking fission products, thyroid-seeking fission products, whole-body-seeking fission products, and so on. Based upon factors such as fission yield, physical and chemical properties, dietary and metabolic requirements of man, biological half-lives, and others, the functional group of outstanding public health significance is the bone-seekers. These are, in particular, the alkaline earth or calcium-like bone-seeking fission products.

Since milk is considered to be the most important single vector of population exposure to calcium-like bone seekers, the Division of Laboratories and Research of the New York State Department of Health has been putting a high priority upon the development of radiochemical procedures for the estimation of calcium-like fission products in milk.

Several procedures for the estimation of Sr^{90} and Sr^{89} were studied but they were all found to be too slow for emergency monitoring of milk supplies and too demanding of time and manpower for intensive monitoring of milk sources and supplies on a state or local health agency level.

The procedures detailed below permit completion of a few samples by a single technician in 6–8 hr when necessary and lend themselves to assembly line operation where large numbers of samples are to be analyzed.

Radiochemical procedure

(1) To 450 g of milk in a 2 l. Pyrex beaker add 100 ml of HNO_3 (conc.) plus 200 mg each of non-radioactive Sr^{2+} and Y^{3+} and approximately 100 c/min (γ) of Sr^{85}.*

(2) Heat to boiling. When foaming subsides, continue to boil for 2 hr keeping volume constant with distilled water.

(3) Cool to room temperature or below, filter through coarse filter paper, wash and discard retained material.

(4) Adjust pH of filtrate to 3 with NH_4OH (conc.), heat to boiling and add slowly and with stirring about 18 g of $(NH_4)_2C_2O_4$. After 10 min of boiling add 3 g of $(NH_4)_2C_2O_4$ and simmer for 30 min more. Then cool to 60°C and add 3 g more of $(NH_4)_2C_2O_4$ with stirring.

(5) Allow sample to cool to room temperature and filter through Whatman #42 filter paper. Wash ppt. with 1% $(NH_4)_2C_2O_4$ solution.

(6) Ignite ppt. and paper at 800°C to a white ash.

(7) Weigh ash to nearest milligram in a tared plastic γ-counting tube graduated to 5 ml.†

Radiometric procedure

(8) Gamma-count sample at from 0.483 to 0.543 MeV vs. prepared standard of Sr^{85} to yield recovery data for Sr isotopes.‡

(9a) Transfer sample to 4π planchet and low background β-counter, and gross β-count for alkaline earth bone-seeker vs. $K^{40}Cl$ standard taken as 914 dis/min per g.§

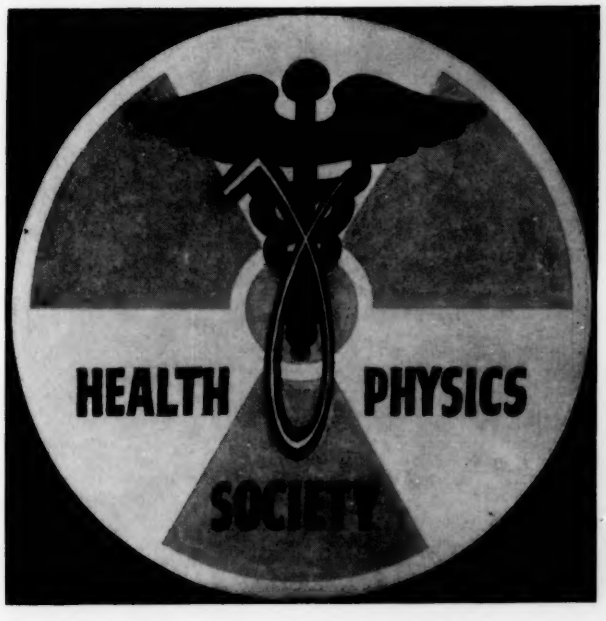
(10a) Dissolve sample in 1:1 HCl and adjust pH to 5 with NH_4OH.

* In this laboratory a single channel γ spectrometer is used to count the Sr^{85}. A channel width of 60 kV is used to monitor the range from 483 kV to 543 kV.

† It has been found convenient to use Lusteroid tubes as supplied by Atomic Accessories, Inc., Jamaica, Long Island, New York.

‡ The comparison count of Sr^{85} is made in the same shaped tube, using very similar geometry and weight of absorber. Thus since the geometry is nearly constant for a volume up to 4 ml, it is only necessary to get the same weight of absorber into the tube as the weight found in step (7).

§ The 4π counter used in this laboratory consists of two pancake tubes (#1001T, Anton Electronics, Inc., Brooklyn, N.Y.), mounted face to face in a lucite frame set into a lead pig. Background is about 35 counts/min. The frame is so arranged that a sample in a planchet may be slipped between the tubes and slots are machined into the lucite frame to accept one absorber above and one below the sample. Using a 1 g sample of KCl which is taken as 914 dis/min, the efficiency of our counting system is 50 per cent. The planchet is simply an aluminum plate with a 2 in. disc out of the center. One side of the plate is covered by 0.0005 Mylar glued to the surface. The sample is placed in the cup so formed and then the other side is covered by the same thickness of Mylar. This planchet accommodates up to 3.5 g of sample.


HEALTH PHYSICS
SOCIETY

(11a) Filter on coarse paper using dilute NH_4OH to wash ppt.

(12a) Repeat (10a) and (11a).

(13a) Ignite paper and ppt. to a white ash.

(14a) Weigh ash to nearest milligram.

(15a) 4π β-count as Y^{90} or β-count in low-background β-counter for Y^{90}.

(9b) Transfer sample to 50 ml quartz centrifuge tube made to fit into large well of tri-carb liquid scintillation β-spectrometer.

(10b) Dissolve sample to minimum volume of 1:1 HNO_3. Add a sufficient quantity of Pilot *B* scintillating beads to take up all the liquid. Cap with plastic cap, insert in tri-carb liquid scintillation spectrometer and β-count vs. $K^{40}Cl$ solution as a standard taken to yield 914 dis/min per g and report as total alkaline earth bone-seeking activity.

(11b) To obtain first approximation of Sr^{90} content, remove quartz tube from well, add 15 ml of H_2O and adjust pH of solution to 5. Mix well.

(12b) Spin in centrifuge to separate $Y(OH)_3$ and decant supernatant liquid.

(13b) To mixture of beads and $Y(OH)_3$ in tube add 1:1 HCl to redissolve ppt., make volume up to 30 ml, adjust pH to 5 with NH_4OH, stir well and spin down beads and ppt. of $Y(OH)_3$.

(14b) Decant supernatant liquid and add to that of (12b). These supernatant liquids may be stored for further Y^{90} "milking".

Division of Laboratories and Research
New York State Department of Health
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N. Irving Sax
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Suggested Insignia for the Health Physics Society

(*Received* 9 *January* 1959)

The two designs (*facing p.* 216) are suggested as suitable for the Health Physics Society insignia. The designs are composed of features which should be familiar to everyone engaged in work involving ionizing radiation. These designs require no elaborate explanation as to what each feature means. The basic feature of each is the radiation symbol which should be the most prominent feature of the Health Physics Society insignia.

A look at some of the insignias in other fields such as, chemistry, geology, etc., reveals that each is relatively simple in design and composed of features which are characteristic of the particular field. What more characteristic symbol do we have in Health Physics than the radiation symbol?

The Martin Company
Baltimore 3, Maryland

P. R. Guinn

Fabrication of Thick Homogeneous β Sources

(*Received* 15 *January*, 1959)

Often, when working with material known to contain β-emitters, it becomes necessary to use thick samples in order to obtain significantly high count rates. When such a problem arose in the analysis of organic samples here at Atomics International, it was found necessary to calibrate the instruments against similar "thick" standards. Liquid β-standards were tried but it was found that they quite often contaminated the counting equipment and their accuracy was not dependable with the passage of time. Because of the nature of some samples which were being studied here, it was essential that thick solid β-standards be constructed for calibration. These standards ideally should have the same geometry and be of the same density (1 g/cm^3) as the unknown samples.

After trying various materials, the problem of fabricating such standards was nicely solved by mixing known amounts of the required radioisotope in a water solution with known weights of a commercially available, water hardening material, called "wood putty"*, which was purchased at a local hardware store. For our purpose it was found that 4 g of this powdered material when mixed with 2 ml of water containing the desired radioisotope produced, upon hardening, excellent solid sources, which we could use for routine calibration.

It was found, interestingly enough, that the water remaining in this material after mixing is rapidly removed by evaporation following a strictly exponential removal with a 4.2 hr half-life until, after about 12 hr, the water content becomes equal to 13.0 $\pm$ 0.2 per cent of the initial powder weight used. This amount of water is assumed to be chemically bonded, for at this point the water content ceases to decrease and becomes constant. Thus, when a known activity is mixed with a known weight of powder to make a thick sample, the activity per gram can be found simply by dividing the total activity by 1.13 times the original powder weight used.

* Schalk's "wood putty", Schalk Chemical Co., Chicago. 351 East 2nd St., Los Angeles 12, California.

It was found that this material gave solid, reproducible, homogeneous β-standards having a density of 1.15 g/cm^3 approximately that of tissue, which could be used for routine calibration without danger of contamination. Sources formed from this material might also be used to advantage in radiation therapy where they could easily be formed around irregular areas requiring treatment to deliver a known dose rate to affected tissues.

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NEWS

News Editor: J. J. FITZGERALD

Health Physics in the News

THE following items concerning Health Physics have appeared in newspapers, public press, or technical journals during the last quarter.

AEC extends deadline for comments on amendments to Part 20

The Atomic Energy Commission extended to 31 August, 1959, the deadline for comments on the amendments on standards for protection against radiation (Part 20). The amendments in general incorporated the latest recommendations of the N.C.R.P., which are discussed in a report by the Commission dated 24 April, 1959. These amendments were published in the Federal Register on 2 May, 1959, and they have been the subject of considerable comment. Hence an extension of time was deemed wise in order to study comments and to integrate these comments into the final proposal.

AEC announces special fellowships for advanced training in Health Physics

On 9 July, 1959, the Atomic Energy Commission announced a new program of special fellowships for advanced training in Health Physics. The program offers five new fellowships per year, beginning with the academic year 1959–60, leading to a doctorate degree in disciplines such as radiation physics, chemistry, biology, and engineering, which are related to and contribute to Health Physics. Appointments will carry a stipend of $4000 per year plus $400 per dependent. Information regarding these fellowships is available from the Oak Ridge Institute of Nuclear Studies, Oak Ridge, Tennessee.

Extensions in radiological physics eductional programs announced by AEC

Changes in presently existing Educational Programs for Radiological Physics have been announced by the Atomic Energy Commission.

Future successful candidates for AEC Radiological Physics Fellowships may be able to choose the University of California, or Harvard, or Michigan, to pursue their Health Physics education. These universities have been added to the present list of four universities available for Radiogical Physics Fellows. Approximately seventy successful candidates will enroll at the Universities of California, Kansas, Rochester, Vanderbilt, and Washington for the 1959–60 academic year.

In addition, under a "Special Assistance Training in Radiation Control" program, several members of state organizations responsible for radiation protection and control have been awarded assistance by the AEC.

Nuclear Reactor Hazard Evaluation Course at Oak Ridge

On 5 May, 1959, the Atomic Energy Commission announced that the Second Session of a Specialized Course in Nuclear Reactor Hazard Evaluation would begin on 2 November, 1959, at Oak Ridge. The course will be open to foreign scientists from friendly nations, and it will seek to develop the ability of the students to evaluate the hazards of reactor operations, including those associated with criticality. The possible hazards range from those involved in the geographical and geological aspects of a reactor site to accidents that might be possible through a chance combination of unlikely events.

Yugoslavian reactor incident—additional data

During a subcritical experiment on 15 October, 1958, the zero energy (heavy water–natural uranium assembly) at the Institute of Nuclear Sciences near Belgrade, Yugoslavia, became uncontrollably supercritical. Six men in the immediate vicinity of the bare reactor received very high doses of neutrons and ionizing radiations. Two other persons, further away, also received doses above the permissible level. It was calculated that the total "average wholebody dose received by the highly irradiated persons from neutron and gamma radiation was 683 rems with a variation of about 15 per cent introduced by different distances from the reactor".

One exposed person died on 15 November because of complications in the respiratory system and the gastrointestinal tract. This death occurred in Paris at the Curie Foundation Hospital, where the heavily irradiated persons had been sent for treatment. Since that time, all the other highly exposed persons responded to treatment and returned to Yugoslavia. The history of their treatment as well as the methods used will be reported by the doctors of the Curie Foundation.

A special committee appointed by the Chairman of the Federal Nuclear Energy Commission (Yugoslavia) examined the causes which led to the accident. Here is a summary of their findings:

(a) During the experiment in question, *none* of the normally used instruments for dosimetry, alarm, and automatic safety control was turned on.
(b) The reactor became supercritical because of the uncontrolled rise of the heavy water level in the reactor tank.
(c) The radiation monitoring equipment and the automatic release of safety rods were neither designed nor constructed as an interlock system, so that it was possible to put the reactor in operation even if such equipment were removed or turned off.
(d) Under such circumstances, in the absence of signalling equipment and automatic shut-down of the reactor, the personnel first noticed an abnormal condition only by the smell of ozone formed in the air.
(e) The energy produced by the reactor during the supercritical state was calculated to be approximately 8×10^7 watt-sec.
(f) The total neutron dose was calculated to be approximately 388 rems, and the gamma dose 295 rems, for an "average" total of 683 rems.

Study of sea-buried radioactive waste

The Coast and Geodetic Survey and the AEC announced recently that a study of simulated radioactive waste packages will be conducted this summer by skindivers in 50 ft of water off the coast of Martha's Vineyard, Massachusetts. Waste containers will be filled with a colored dye which will be readily recognized in the water if the packages leak. The skindivers, using underwater color cameras, will photograph the containers as they descend. The dye studies are part of a larger project planned to find out what happens to radioactive waste at sea.

Progress in plutonium ingestion therapy

Dr. JACK SCHUBERT, a senior chemist at Argonne National Laboratory, recently reported that his laboratory research team has had success with two chelating agents in flushing out plutonium from the bodies of laboratory experimental animals, even though the plutonium had been retained in tissues for *several days*.

The two substances, known as DTPA and BAETA, sequester or trap metal ions to form a stable, water soluble complex that is relatively chemically inert. The complex is flushed out of the body by normal elimination processes. In one experiment, rats were given a single injection of DTPA and BAETA 6 *days* after exposure to plutonium. Within 24 hr the rats excreted more than 5 per cent of the plutonium retained in their bodies. This is about 150 times as much as they would have eliminated without treatment.

Previously some substances (notably zirconium citrate and EDTA) have proved effective in removing plutonium, provided treatment was delayed no longer than 8 *hr* after exposure.

AEC statement on fallout

Excerpts quoted below are from a statement to the AEC by the General Advisory Committee to the AEC concerning problems presented by radioactive fallout (dated 4 May, 1959):

"It is now apparent that the circulation of the upper atmosphere, and particularly the stratosphere, is much more complicated and the concentration of bomb debris less uniform than had been anticipated when early estimates (of fallout) were made. This has resulted in non-uniform distribution of fall-out with higher concentrations in the middle latitudes of the Northern Hemisphere.

"A reasonable estimate of the amount of fission products that has been injected into the *stratosphere* by all nuclear tests is 65 megatons (TNT equivalent) of fission energy. This corresponds to about 100 lb of Sr^{90} in the entire stratosphere. It is estimated that fully 50 per cent of this Sr^{90} has already fallen out.

"In order to place the hazard of fallout in proper perspective, it should be pointed out that the amount of *total body external radiation* resulting from fallout to date, together with future fallout in any part of the world from *previous* weapon tests, is:

(a) less than 5 per cent as much as the average exposure to cosmic rays and other background radiation.
(b) less than 5 per cent of the estimated average radiation exposure of the American public to X-rays for medical purposes."

"In regard to internal effects of Sr^{90} from ingestion, the amount of Sr^{90} which has been found in food and water is less of a hazard than the amount of radium normally present in public drinking water supplies in certain places in the U.S. and in public use for many decades."

AEC curtails importation of Sr^{90} watch dials

In April of this year, the AEC issued "regulatory orders" to three American watch companies to halt further importation and sale of luminescent watch dials containing Sr^{90}. The three companies reportedly denied knowledge of the presence of strontium as the luminescent agent.

IAEA to build atomic laboratory

The International Atomic Energy Agency recently approved plans for the construction of an atomic laboratory near Vienna. Said IAEA: "The laboratory will serve for the standardization of isotopes and preparations of radioactive standards; calibration and adaptation of measuring equipment; quality control of special materials for nuclear technology; measurements and analysis in connection with the Agency's safeguards and health and safety program and services for member states." The laboratory is planned to occupy a site adjacent to the Austrian Organization for the Study of Atomic Energy reactor and laboratory facilities now under construction.

British plan nuclear medical center

The British Royal Marsden Hospital announced recently that it will build at Sutton Downs, Surrey, the first hospital in Britain devoted solely to the application of nuclear energy to medicine. A 35 MeV linear accelerator for treatment of cancer patients is planned. Study and development of the use of radioisotopes in medicine will be a major part of the research program.

Radiological health problems—most important emerging public health program

The American Public Health Association has reported the findings of an eighty-member national panel of federal, state and local public health directors. The most important emerging public health problem was considered to be the radiological health problem. The *control* of radiological health was rated as the *second* outstanding public health achievement in 1958 (polio rated first).

Steel drum with radiation markings found on Oregon beach

On Saturday, 4 July, 1959, a steel drum was found on a beach at Battle Rock State Park, Oregon, marked in a manner indicating radioactive contents. Examination of the drum and its contents showed no sign of radioactivity.

A number of questionable points were noted:

(a) There were no identifying marks on the barrel as required of authorized waste disposal organizations.

(b) The drum was painted with white enamel, whereas commonly used drums are not so painted.

(c) The condition of the enamel indicated only relatively short exposure to the elements.

(d) The drum was not weighted with concrete to insure sinking.

(e) The markings on the drum were not those customarily used in waste disposal activities.

Kellogg radiation suit

Recently two former employees of the M. W. Kellogg Company who had been exposed to radiation in an accident involving the dispersal of radioactive Ir^{192} in March 1957, at the company's South Houston, Texas, plant, filed suit against the Phillips Petroleum Company for $426,635 in damages. They maintained that Phillips, as operator of the Materials Testing Reactor where the pellets were irradiated, was responsible for the accident, which, they assert, has been the cause of a serious decline in their health in the past two years. A 1957 AEC report said that an investigation of the mishap had shown no evidence of radiation damage to the plant's employees.

Tests to determine shielding provided by homes from fall-out

The AEC announced in July that tests had commenced in the city of Oak Ridge, Tennessee to determine the shielding provided by homes against gamma radiation from simulated fall-out. Personnel from the Health Physics Division of the Oak Ridge National Laboratory are conducting the experiments. These studies constitute an extension of measurements at the Nevada Test Site in 1958 in which several unoccupied houses had been exposed to the radiation from several hundred small Co^{60} sources. The Oak Ridge experiments are intended to provide data for fully furnished homes located on various types of terrain.

Health Physics Society Meeting 1959

The 1959 Annual Meeting of the Health Physics Society was held at the Civic Auditorium in Gatlinburg, Tennessee on June 18, 19 and 20. There were 552 registrants. This exceeded the attendance of any previous meeting. The annual banquet was held on June 19 with 376 attending. The banquet speaker was Dr. Andrew Holt, President of the University of Tennessee. Tours of the Oak Ridge National Laboratory and of the Smoky Mountains were available to the participants as well as an afternoon of foreign technical films. The wives of the members were treated to three days of planned ladies' activities. There were 24 commercial exhibits covering all phases of Health Physics activities. The technical aspects of the meeting are covered elsewhere. The abstracts of the papers presented at the Meeting have appeared in the Journal* and many of the papers will also be published. The meeting can be summarized as a success—technically, socially and financially.

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* *Health Phys.* **2,** 87 (1959).

Scientific exhibits for Industrial Health Conference

Applications are being solicited for scientific exhibits for the 1960 Industrial Health Conference which will be held in Rochester. Exhibits will be shown from 26 April to 28 April. Application blanks and information may be obtained from GEORGE A. HARDIE, M.D., Chairman of Scientific Exhibits Committee, 343 State Street, Rochester 4, New York.

A Scientific Conference on the Disposal of Radioactive Wastes

This is to be held from 16 to 21 November 1959, in the Principality of Monaco, under the joint sponsorship of the International Atomic Energy Agency (IAEA) and the United Nations Educational Scientific and Cultural Organization (UNESCO), with the co-operation of the Food and Agriculture Organization of the United Nations (FAO). The program covers such radioactive wastes as result from the peaceful uses of atomic energy and envisages problems arising from their possible disposal into the sea and on land. The meeting will be conducted in English, French, Russian and Spanish. The abstracts (about 500 words) and the papers (less than 6000 words) should be submitted in one of these four languages. The abstracts and papers should be transmitted to the Secretariat as early as possible and not later than October 1959. It is recommended that communications be directed through the Ministry of Foreign Affairs or other appropriate government channels in each country, to permit effective co-ordination of planning.

Notice from the IAEA

A need is rapidly developing for experienced health physicists to assist countries in underdeveloped areas in setting up their health physics services. A wide range of experience, both technical and administrative is called for, as in most cases the assistance needed covers such diverse subjects as setting up a national film dosimeter service, advice and controls for the use of radiosotopes for medical and other purposes, protection from research reactors, waste disposal, environmental contamination studies and supporting training programes.

The need to be able to provide sound advice based on experience in such a variety of fields when working under rather isolated conditions will naturally limit the availability of candidates. It is, however, hoped that candidates who are weak in one or two aspects can be supported by special briefing and the help of additional visiting specialists.

The period for which assistance is required while local technical and scientific personnel are acquiring the experience needed, is estimated to range between six months and two years.

Health physicists who are interested in such appointments are requested to inform the Health, Safety and Waste Disposal Division, International Atomic Energy Agency, Vienna I, Austria, who will be pleased to supply further information.

A LIST OF PAPERS TO BE PUBLISHED IN FUTURE ISSUES

C. W. SILL and J. K. FLYGARE, JR.: Iodine monitoring at the National Reactor Testing Station.

R. H. GOEKE and C. L. WEAVER: Health physics field operations in Atomic Energy Commission weapons testing program.

A. L. BONI: Urinalysis method for enriched uranium.

R. L. BLANCHARD, B. KAHN and R. D. BIRKHOFF: The preparation of thin, uniform, radioactive sources by surface adsorption and electrodeposition.

S. M. SANDERS, JR.: Power functions relating excretion to body burden.

P. W. DURBIN: Metabolic characteristics within a chemical family.

G. W. ROYSTER, JR.: Electrodeposition of uranium from urine.

R. E. COFIELD: *In vivo* gamma counting as a measurement of uranium in the human lung.

H. FOREMAN, W. MOSS and W. LANGHAM: Plutonium accumulation from long-term occupational exposure.

H. MUTH, B. RAJEWSKY, H. J. HANTKE and K. AURAND: The normal radium content and the Ra^{226}/Ca ratio of various foods, drinking water and different organs and tissues of the human body.

T. TAMURA and D. G. JACOBS: Structural implications in cesium sorption.

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METABOLIC CHARACTERISTICS WITHIN A CHEMICAL FAMILY*

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(*Received* 4 *June* 1959)

Abstract—The available data have been reviewed for the biological behavior in rats of high specific-activity radioisotopes of seventy elements. Tracer quantities were administered in single intramuscular injections. Groups of rats were autopsied at various intervals, and tissues and excreta were assayed for radioactivity.

Distribution data were arranged according to the grouping of the periodic table of the elements. The anions (including the halogens), the oxygenated or halogenated ions of Groups IV, V and VI, and the transition metals were rapidly eliminated by the kidney. The monovalent alkali metals were distributed almost uniformly in soft tissue with subsequent excretion by the kidney. The bivalent cations, except Hg^{2+}, Cd^{2+} and UO_2^{2+}, were deposited primarily in bone mineral and were eliminated slowly in both urine and feces. The tripositive elements of Group III, the lanthanides, and the actinides were deposited in liver and bone. The liver fraction was excreted via the bile without recirculation, while that deposited in bone was turned over at a rate slower than that of normal bone remodelling. The quadrivalent cations such as Zr^{4+}, Th^{4+} and PuO_2^{2+} were deposited almost exclusively in bone and were bound more strongly than the Group III elements.

The properties that determine biological behavior are: (a) the oxidation state stable at body pH; (b) the solubility of the stable state; (c) the tendency to be incorporated into organic compounds; and (d) the tendency to associate with specific proteins.

INTRODUCTION

THE great potential hazard of the radioactive products of nuclear fission and of the fissionable elements was recognized almost as soon as the nuclear chain reaction was successfully demonstrated. The necessity for strict control measures was apparent. To provide a sound basis for such control measures an intensive investigation was begun of the metabolic behavior and biological effects of these radioactive substances. By 1946, a wealth of information had been gathered from these radiobiological researches which provided a logical framework for the establishment of minimum precautions to be taken by users of radioactive materials and for the protection of the general public.[1–5] A great deal of ground remained to be covered. With the increasing use of radioactive substances many radionuclides, in addition to the fission products and the heaviest elements, would soon be produced in reactors and cyclotrons in hazardous quantities. Under the direction of the late Dr. J. G. HAMILTON, a systematic investigation was made of the biological behavior in laboratory rodents of tracer amounts of some seventy radioelements. Most of the radioisotopes were prepared on the Crocker Laboratory 60-in. cyclotron by Dr. T. M. PUTNAM and Mr. G. B. ROSSI and their staff. Dr. W. M. GARRISON and his co-workers isolated and purified the radioactive products and prepared them for biological use. Dr. K. G. SCOTT directed the animal work. This report is derived largely from the data collected by this group during the years 1943 to 1952.

* This work was performed under the auspices of the U.S. Atomic Energy Commission.

Data for several elements that were not investigated in this laboratory have been drawn from the literature.

Some of the data contained in this summary report have been published elsewhere in various forms.[1,5–16] They have been reanalyzed for this presentation and information is included that was previously available only in University of California Radiation Laboratory Project Reports. The distribution and elimination data have been arranged to compare the biological behavior of the elements of the various periodic groups.

Since Mendeléeff's presentation of the periodic table, the chemical properties of elements have been successfully predicted from knowledge of the behavior of other members of the same periodic group. The properties of the other halogens, for example, suggested the basic analytical methods first used to isolate and identify element 85, astatine.[17] In many instances the biological behavior of an untested element has also been predicted (at least qualitatively) from knowledge of its near chemical neighbors. It was logical to look for, and not surprising to find, halogens other than iodine concentrating in the thyroid gland,[18] and to find alkaline earths other than calcium being incorporated into bone mineral.[19]

Meaningful comparisons of the behavior of the elements of each periodic group and subgroup were possible because the tracer experiments were performed by the same group of experimenters, using the same analytical techniques and equipment, and the same animal species as the test system.

METHODS

Detailed descriptions of the preparation of the radioisotopes and of the biological procedures appear elsewhere,[6,7,19a] and only a brief outline of the experimental methods is presented here. The radioisotope of each element investigated was selected on the basis of ease of preparation in carrier-free form or at very high specific activity, length of half-life, and desirable radiation characteristics. The specific radioisotopes used are shown in the Tables of data.

The animals used were mostly female adult albino rats. Radioisotopes of the elements presumed to be anions or monovalent cations were injected intramuscularly or intravenously in isotonic saline. Radioisotopes of cations in the di-, tri-, or quadrivalent state were injected intramuscularly, usually in isotonic or hypertonic sodium citrate. The complexing agent was used to prevent radiocolloid formation either in the injection solution or in the animal's circulation, and to facilitate absorption from the site of injection. A minimal amount of each radionuclide was administered to avoid radiation effects, but enough was given to ensure accurate radioactive assay of tissues and excreta at the longest time-interval studied. After injection, groups of three rats were placed in metabolism cages for the separate collection of urine and feces. Cages of three rats were autopsied at intervals varying from 1 hr to 8 months, the length of each experiment depending upon the half-life of the isotope and the quantity available.

Tissues and excreta were usually oven dried at 100°C, ashed in a muffle furnace at 500°C, and dissolved in dilute nitric acid. Aliquot samples were evaporated in small porcelain capsules and assayed for β-particle activity with a thin window GM counter. A known portion of the injection solution served as a counting standard, and self absorption curves were prepared for each radioisotope by the dilution method. In some cases γ-rays were sufficiently intense to permit accurate assay with a lead-shielded GM tube. After 1951 a NaI–Tl crystal scintillator was used for γ-ray counting. As, Tc, Se, Te, I and Ge were found to be partially volatile at muffle-furnace temperatures. These radioelements and those with very short half-lives (At, Sc and La) were either dried at temperatures less than 100°C, and the β-particle activity measured; or fresh tissues were assayed for γ-activity when γ-rays were present at sufficiently high intensity. In all cases samples were counted for a long enough time to keep counting errors within the range of ± 5 per cent. A balance sheet of radioactivity was prepared for each animal in every experiment. Samples were rechecked, or the experiment was repeated until the total recovery varied by no more than ± 10 per cent from the amount injected.

The method of calculating the radioisotope distribution in terms of per cent of administered dose (per cent of absorbed dose in oral or intramuscular experiments) has already been described.[6]

Several of the lightest elements appear as blanks in the tables. Some—oxygen, silicon, boron, aluminum, nitrogen, lithium, magnesium and chlorine—either could not be prepared carrier-free or have only very short-lived radioisotopes. Comparable tracer data in the rat were not available for several other elements: sulfur, carbon, titanium and nickel.

The chemical states of the elements as they appear in the tables were not determined experimentally,

but rather are best estimates based on the chemistry of separation from the target material and a study of the oxidation-reduction potentials.[20]

The tracer data are collected in Tables 1 through 10. In most cases the 4-day postinjection interval is shown because data were available for the majority of the elements at this time period. Results of shorter-term tracer studies are shown for those elements that were very rapidly eliminated or for those with half-lives shorter than a few hours. Several biological criteria suggested the selection of the 4-day interval as most informative: (a) intramuscularly injected material had been largely absorbed—less than 10 per cent remained at the site of injection when a complexing agent was added; (b) the blood stream had very nearly been cleared: (c) tissues whose initial radioisotope concentrations represented blood-borne isotope or simple mixing with extra-cellular fluid had been cleared; (d) prompt excretion, reflecting initial clearance of the blood had very nearly ceased; (e) the excretion resulting from slower processes of turnover had not yet become important.

In the tables, radioisotope content is shown for those tissues which are important sites of deposition for at least one member of the particular periodic group. The symbol *U* or *F* to the right of the value for total excretion indicates that the greater proportion of a particular radioisotope was eliminated by the kidney (*U*) or by the digestive tract (*F*). Where the symbol *UF* appears, about equal amounts were eliminated by the two routes.

GROUP I: THE ALKALI METALS

As shown in Table 1, skeletal muscle was the important deposition site of most of the Group I elements. The kidney was the chief route of elimination. A few days after injection the concentration of these metals in muscle was three to four times that of the blood and one and one-half times greater than the concentrations in other soft tissues. The behavior of the group was not completely uniform; Na tended to be eliminated almost twice as fast as the other members of the group, and Na and Rb concentrated in bone to a significantly greater extent than K or Cs.

The metabolic behavior of the Sub-group I elements—Cu, Ag and Au—differed from the elements of the main group. Incomplete data of ASHIKAWA *et al.*[21] indicate that in adult rats Cu^{2+} concentrated in the liver, kidney and spleen. Tracer data were not available for bone; however, the values of TIPTON *et al.*[22] for trace elements in human tissues show bone concentration of Cu to be about 10 per cent of the concentration in the soft tissues.

Silver in the 1+ state has a unique fate. SCOTT and HAMILTON[9] found that injected carrier-free radiosilver combined almost quantitatively with plasma protein, was rapidly removed from the circulation by the liver, and

Table 1. *Distribution of radioisotopes of the elements of Group I, the alkali metals, in various tissues of the rat after intramuscular injection*

Radioisotope	Chemical form administered	Time	Percentage of absorbed dose				Ref.
			Bone	Liver	Muscle	Excreta	
Li	—	—	—	—	—	—	—
Na^{22}	NaCl	2 days	6.1	1.3	10.3	55.2 *U*	25
$K^{42,43}$	KCl	2 days	4.2	3.3	47.6	26.9 *U*	26
Rb^{86}	RbCl	4 days	14.4	4.8	43.6	20.8 *U*	27
$Cs^{134,135}$	CsCl	4 days	1.5	2.1	38.5	44.9 *U*	4
Sub-group I							
Cu^{64}*	$CuCl_2$	6 hr	—	2.9	0.1	—	21
Ag^{110}	$AgNO_3$	1 day	4.2	2.8	2.2	60.9 *F*	9
Au^{196}†	$Au_2(S_2O_3)_3$	1 day	6.9	66.4	2.3	6.8 *U*	28

* Kidney 10 per cent, spleen 6.2 per cent.

† Injected intravenously.

Table 2. Distribution of radioisotopes of the elements of Group II, the alkaline earths, in various tissues of the rat after intramuscular injection

Radioisotope	Chemical form administered	Time (days)	Percentage of absorbed dose				Ref.
			Bone	Liver	Kidney	Excreta	
Be^{7}	$BeCl_2$	4	32.2	9.6	2.0	49.0 *U*	10
Mg	—	—	—	—	—	—	—
Ca^{45}	$Ca_3(Cit)_2$	4	70.0	—	—	30.0 *F*	34
Sr^{90}	$Sr_3(Cit)_2$	4	44.0	—	—	56.0 *UF*	34
Ba^{140}	$BaCl_2$	4	37.3	—	—	55.0 *UF*	16
Ra^{223}	$Ra_3(Cit)_2$	4	40.4	—	—	59.6 *UF*	36
Sub-group II							
Zn^{65}*	$ZnCl_2$	1	—	38.5	2.4	—	35
Cd^{109}	$CdCl_2$	4	3.3	77.8	6.1	5.6 *F*	7, 27
$Hg^{196,197}$	$HgCl_2$	5	1.2	7.9	25.5	49.0 *F*	7, 37

* Tumor-bearing mice data.

was eliminated by the gastrointestinal tract via the bile. In an attempt to study the fate of a presumably soluble Au compound, radioactive auric thiosulfate was prepared. After intravenous injection of this compound colloidal aggregates were apparently formed. The distribution pattern was typical of colloids of small particle size, that is high concentrations of radiogold were found in liver, spleen and bone marrow.[23,24] The behavior of non-colloidal Au remains to be demonstrated.

GROUP II: THE ALKALINE EARTHS

A summary of the tracer experiments with the alkaline earth elements is shown in Table 2. The metabolic behavior of these elements was nearly uniform in the rat. After the first few days more than 95 per cent of retained Ca, Sr, Ba or Ra was found in bone. Tracer data are not available for Mg, but TIPTON's spectrographic analysis of human tissue shows that bone contains ten times more Mg per unit weight than does any other tissue—a value that agrees well with analyses for the other alkaline earths.[22] With the possible exception of Be, the alkaline earth elements in bone are associated with the mineral phase. Calcium receives preferential treatment by the kidney[29] and by the gastrointestinal tract,[30] so that after oral administration or parenteral injection a greater percentage of a single dose of radiocalcium is deposited in the skeleton than any other element of the group.

Recent evidence indicates that the alkaline earth in mineral bone is distributed in at least three grossly observable compartments—one with a rapid turnover, one of intermediate half-time, and a third that is turning over very slowly. It has been postulated that these three compartments represent (a) rapid physicochemical exchange, (b) gross bone turnover due to growth and (or) remodeling,[31] and (c) very slow exchange processes resembling solid state diffusion.[32] The amount of administered alkaline earth tracer that appears in the various turnover compartments depends largely on the age of the animal; the older the animal, the smaller the percentage that will be associated with the slower turnover compartments.[33] In rats the half-times of the various skeletal turnover compartments have been found to be similar for Ca and Sr.[34] Beryllium concentrated to a significant degree in liver and kidney as well as in bone but was removed from bone at an apparently slower rate than the other members of the main group. Beryllium thus resembles Zn in exhibiting certain of the characteristics of both the alkaline earths and the elements of Sub-group II.

Although apparently in the 2+ state in neutral aqueous solutions, the Sub-group II elements (Zn, Cd and Hg) were not deposited in bone to the great extent noted for the elements of the main group. Immediately after injection, tracer Zn was most highly concentrated in the liver.[35] TIPTON's study of trace elements in human tissues indicates, however, that concentrations of stable Zn in the prostate gland and in bone, are more than twice those in liver, kidney or other soft tissues.[22]

Tracers of Cd and Hg were deposited preferentially in the liver and kidney. Retention of tracer Cd in both organs was quite prolonged.[7] The short half-lives of the $Hg^{196,197}$ isotopes precluded long-term study, but during an 8-day observation period the renal concentration actually increased, while that in the other tissues declined which indicates a high degree of retention.[7] The prolonged retention of Cd and Hg strongly suggests some stable combination of these two elements with protein in the liver and kidney.

GROUP III: THE LANTHANIDES AND ACTINIDES

The distribution of the lanthanides, actinides, and elements of Group III is summarized in Tables 3, 4 and 5, respectively. Two of these elements (Tl and U) resembled other periodic groups in their biological behavior. The stable state of Tl in neutral aqueous solutions is Tl^{+}, and its behavior, i.e. nearly uniform distribution in the soft tissues and rapid elimination, was reminiscent of the Group I elements.[7,38] Uranyl ion, UO_2^{2+}, resembled Hg^{2+} and Cd^{2+} in its tendency to concentrate in the kidney[7,39] and should probably be classified with the elements of Sub-group II.

The behavior of the trivalent cations was remarkably uniform. Liver and bone are the major deposition sites. For those elements deposited primarily in the liver, the gastrointestinal tract was eventually the main route of elimination. For those concentrating primarily in bone, the kidney was the major excretory organ. Skeletal muscle accumulated significant amounts of Ga, In and Sc, the lightest members of the group.

There is some evidence pointing to combination of these highly charged ions with protein in the liver. That fraction of a trivalent ion initially deposited in the liver was eliminated with a half-time ranging from 10 to 20 days. Comparison of the amount originally accumulated in the liver, and the total fecal excretion during the first 60 days postinjection indicated that the elimination pathway was directly from the liver into the small bowel via the bile with little gastrointestinal reabsorption. Retention of trivalent cations in the skeleton was prolonged. The half-time for removal of these elements was

Table 3. Distribution of radioisotopes of the elements of Group III in various tissues of the rat after intramuscular injection

Radioisotope	Chemical form administered	Time (days)	Percentage of absorbed dose				Ref.
			Bone	Liver	Muscle	Excreta	
B	—	—	—	—	—	—	—
Al	—	—	—	—	—	—	—
Ga^{67}	Ga (citrate)	4	19.6	7.1	13.2	48.7 *F*	40
In^{114}	$InCl_3$	4	16.8	14.0	14.7	25.8 *F*	7, 41, 67
$Tl^{200,202}$	TlCl	4	5.4	2.6	28.5	50.1 *F*	7, 38
Sub-group III							
Sc^{46}	Sc (citrate)	4	15.9	21.2	12.5	31.1 *F*	28
Y^{91}	YCl_3	4	55.6	12.1	2.6	26.3 *U*	42
La^{140}	La (citrate)	4	18.4	64.2	1.2	11.3 *UF*	6

*Table 4. Distribution of radioisotopes of the rare earth elements, the lanthanons, in various tissues of the rat 4 days after intramuscular injection as complex citrates**

Radioisotope	Percentage of absorbed dose			
	Bone	Liver	Urine	Feces
Ce^{144}	27.7	51.0	6.0	8.0
Pr^{143}	26.6	48.4	6.6	8.6
Nd^{147}	31.2	27.1	22.1	10.0
Pm^{147}	36.4	41.4	9.6	6.5
Sm^{153}	33.2	34.8	12.7	13.1
$Eu^{152,154}$	35.6	25.0	16.6	11.1
Gd^{159}	41.4	12.1	26.9	10.1
Tb^{160}	60.5	6.8	15.6	6.9
Dy^{166}	59.9	2.8	24.1	6.2
Ho^{166}	55.6	2.4	21.2	13.2
Er^{169}	56.4	1.2	27.4	7.4
Tm^{170}	64.1	1.9	22.1	5.2
Yb^{175}	57.8	2.6	19.3	7.3
Lu^{177}	67.6	2.7	15.6	7.3

* See Ref. (6a) for complete tabulation of data.

2.5 years on the average for the rat. While gross autoradiographs indicate that these elements concentrate on bony surfaces and at sites of resorption and growth,[5,11–13,16,47–49] the question of the ultimate skeletal deposition site—the surface of the organic matrix or the bone salt—is yet to be completely resolved.

The more basic light lanthanons with their large ionic radii tended to be deposited largely in the liver. The heavier lanthanons, more acidic and of smaller ionic size, were laid down exclusively in bone. Yttrium and the actinides whose stable oxidation state is greater than 3+—Pu, Np, Pa and Th—resembled the heavy lanthanons, while the tripositive actinides—Ac, Am and Cm—behaved like light lanthanons.

GROUP IV

The important deposition sites of tracers of the Group IV elements are shown in Table 6. These elements constitute the third major group of bone seekers. There was one demonstrated exception, Ge, which under physiological conditions exists in hydrated anionic form. Tracer Ge was readily absorbed from an intramuscular injection site and was rapidly and almost uniformly distributed in the soft tissues. It was almost quantitatively excreted by the kidney in the first 24 hr after injection. After a few hours the concentration in bone was somewhat greater than that in most of the other tissues, but this deposit was quickly eliminated, and within 4 days after injection constituted only 0.5 per cent of the administered dose.

Lead has long been recognized as a hazardous bone seeker. The association of Pb^{2+} with the mineral phase of bone is well documented.[50] Tracer Pb was eliminated by the kidney and

Table 5. Distribution of radioisotopes of the actinide elements in various tissues of the rat after intramuscular injection

Radioisotope	Chemical form administered	Time (days)	Percentage of absorbed dose				Ref.
			Bone	Kidney	Liver	Excreta	
Ac^{227}	Ac (citrate)	4	26.8	0.6	56.4	10.6 *UF*	43
Th^{237}	Th (citrate)	8	66.3	3.3	4.1	14.8 *UF*	44
Pa^{230}	*	4	45.1	4.0	7.8	24.6 *F*	45
U^{230}	UO_2Cl_2	4	10.9	11.8	0.2	76.0 *U*	39
Np^{237}	†	1	44.4	2.6	8.5	36.9 *U*	46
Pu^{239}	PuO_2Cl_2	4	70.9	1.8	8.4	8.2 *F*	11
Am^{241}	$AmCl_3$	4	19.1	2.3	35.7	34.7 *F*	12
Cm^{242}	$CmCl_3$	4	29.0	1.6	40.2	20.0 *F*	13

* Chemical form unknown but oxidation state probably 4+ or 5+.

† Administered in conc. NH_4Cl and Na_3 citrate; oxidation state 4+ or 5+.

Table 6. Distribution of radioisotopes of the elements of Group IV in various tissues of the rat after intramuscular injection

Radioisotope	Chemical form administered	Time (days)	Percentage of absorbed dose Bone	Liver	Kidney	Excreta	Ref.
C	—	—	—	—	—	—	—
Si	—	—	—	—	—	—	—
Ge^{71}	$NaHGeO_3$	4	0.4	0.5	1.1	98.5 *U*	27
Sn^{113}	Sn (citrate)	4	29.4	1.0	3.1	61.2 *U*	7, 42
Pb^{203}	$PbCl_2$	4	27.7	1.9	3.1	62.0 *UF*	7, 53
Sub-group IV							
Ti	—	—	—	—	—	—	—
Zr^{95}	Zr (citrate)	4	34.9	6.6	4.3	17.8 *F*	27
Hf^{181}	NaHf mandelate*	4	15.4	37.8	1.5	5.0 *UF*	52

* Intravenous injection.

gastrointestinal tract in nearly equal proportions, also reminiscent of the Group II elements.

Tin and Zr tracers were deposited in the skeleton to the same extent—30 per cent to 35 per cent of the absorbed dose. Timed studies were not sufficiently long to establish skeletal removal rates; however, from 2 to 4 months after administration nearly two-thirds of the initial deposit remained, indicating skeletal turnover times in excess of 400 days. There is some difference in the manner in which radioactive Sn and Zr were eliminated. A large fraction of the tracer Sn was excreted by the kidney in the first 24 hr. After the first day, urinary excretion became almost negligible, and the intestinal tract took over as the chief excretory organ. Very little carrier-free Zr^{95} was excreted during the first day. In the ensuing months 80 per cent of the eliminated Zr was found in the feces. The early urinary excretion of Zr can be greatly enhanced by the simultaneous administration of milligram amounts of stable zirconium citrate.[51] Presumably colloidal aggregates are formed that are readily filterable by the renal glomeruli, yet are small enough to escape filtration from the circulation by the reticuloendothelial system.

The data shown for Hf are drawn from KITTLE *et al.*[52] Most Hf compounds are relatively insoluble in neutral solutions at moderate temperatures. The high concentration of radiohafnium in the liver does not fit with the deposition patterns of the other members of the group, and suggests that a portion of the sodium–hafnium–mandelate complex was in an aggregated form. The presence of 15 per cent of the administered Hf in bone does indicate a behavior similar to Sn, Zr and Pb.

GROUP V

The distribution of the Group V elements is shown in Table 7. With the exception of Bi and perhaps Nb, the principal oxidation state for this periodic group is 5+. Radioarsenate and, to a small degree, radioantimonate were incorporated into the circulating erythrocytes. During *in vitro* incubation, radiophosphate is believed to exchange with the stable phosphate of the cells, whereas tracer As and Sb apparently form stable compounds with the protein, globin. The cells thus tagged retain the radioactive label until their destruction. The circulating blood was the only important deposition site for both of these radioelements, and much of the apparent tissue deposition of both could be accounted for by blood contained in the tissues at autopsy. That fraction of both isotopes not initially associated with the red cells was excreted in the first few days, chiefly by the kidney. The mean life of As^{74} in the body was

Table 7. Distribution of radioisotopes of the elements of Group V in various tissues of the rat after intramuscular injection

Radioisotopes	Chemical form administered	Time (days)	Percentage of absorbed dose						Ref.
			Muscle	Bone	Liver	Kidney	Blood	Excreta	
N	—	—	—	—	—	—	—	—	—
P^{32}	Na_2HPO_4	1	27.4	17.7	—	*	—	22.8 *U*	54
As^{74}	NaH_2AsO_4	4	1.4	2.3	2.4	0.8	44.1	45.7 *U*	8
$Sb^{122,124}$	$HSbO_3$	4	0.1	0.9	0.1	<0.1	2.0	96.5 *U*	41
Bi^{206}	BiOCl or BiO(OH)	4	0.6	1.5	6.6	14.4	<0.1	75.2 *U*	55
Sub-group V									
V^{48}	$Na_2H_2V_6O_{17}$	4	5.0	9.9	6.2	4.4	1.5	63.9 *UF*	56
Nb^{95}	Nb (citrate)	4	8.0	16.2	8.4	2.9	7.7	39.4 *UF*	7, 45
Ta^{182}	Ta_2O_5 in Na_3 (citrate)	4	12.2	11.9	7.4	5.4	1.2	48.6 *U*	7, 57

* Viscera contained 23.9 per cent of the injected P^{32}.

roughly equivalent to the red-cell life span which indicates that there was little tendency for recirculation of radioarsenic after the destruction of the original host cell.

The tracer data for Bi^{206} agree with pharmacological observations of the distribution of stable Bi compounds. The metabolism of radiobismuth in the 3+ state was not like the majority of the trivalent elements but more closely resembled UO_2^{2+}, suggesting that Bi(III) was in an oxygenated or "basic" form. Like UO_2^{2+}, retention of Bi in the kidney was not prolonged, and by the seventeenth day after injection only 0.6 per cent remained in the kidney, and 95 per cent had been excreted.

The Sub-group V elements tended to combine with the serum proteins rather than with the red cells, and their plasma concentrations were still appreciable as long as 2 weeks after injection. Shortly after injection all three elements—V, Nb and Ta—were distributed throughout the body, with the highest concentrations in the liver, kidney and bone. Initial excretion was chiefly urinary, but after the first few days some was eliminated by the gastrointestinal tract as well. Retention was generally more prolonged in the skeleton than in the soft tissues, and by the second month bone was the critical organ for V and Nb. Skeletal turnover of these elements seemed to be more rapid than the turnover found for the alkaline earths or for the lanthanides.

Vanadium, the lightest of the group, forms the most soluble compounds. It was rapidly and almost completely absorbed and was eliminated nearly quantitatively in the first 2 months. By the sixty-fourth day muscle, skin and liver contained 4.3 per cent of the injected dose, and 4.6 per cent remained in the skeleton.

Niobium belongs to a group of elements with high positive charge—Pu, Tm and Y—that combine in a stable fashion with the serum proteins and remain in the circulation for many hours after injection.[51] Compounds of Nb are less soluble and more basic than those of V, and a complexing agent was required to facilitate absorption. Retention of Nb^{95} was more prolonged than that of V. As long as two months after injection, 28 per cent of the administered dose was retained, with significant quantities in the muscle and skin as well as in liver, kidney and bone.

Compounds of Ta are generally inert and exceedingly insoluble. Only 15 per cent of intramuscularly administered radioactive Ta_2O_5 was absorbed when no complexing agent was added, and intravenously administered Ta_2O_5 was almost entirely colloidal. Like Nb and V,

Table 8. *Distribution of radioisotopes of the elements of Group VI in various tissues of the rat after intramuscular injection*

Radioisotopes	Chemical form administered	Time	Percentage of absorbed dose					Ref.
			Bone	Liver	Kidney	Blood	Excreta	
O	—	—	—	—	—	—	—	—
S	—	—	—	—	—	—	—	—
Se^{75}	$NaHSeO_4$	1 day	8.1	7.3	3.8	6.1	49.8 *U*	46
$Te^{127,129}$	NaH_5TeO_6	1 day	9.5	5.4	5.4	12.7	61.9 *U*	4
Po^{210}	Unknown*	1 day	2.7	21.0	2.0	19.4	—	58
Sub-group VI								
Cr^{51}	$NaHCrO_4$	1 day	10.6	3.3	1.4	6.6	57.5 *U*	59
$Mo^{93,99}$	$NaHMoO_4$	4 hr	3.6	30.3	2.5	2.5	33.0 *U*	7, 55
W^{181}	$NaHWO_4$	1 day	1.2	0.1	0.4	0.2	96.6 *U*	7, 28

* Administered orally, converted to whole tissue estimates from specific activity (%/g wet tissue).

Ta was found in nearly all the tissues, and most highly concentrated in liver, kidney and bone. Tantalum was retained more strongly than the other members of the sub-group, and 8 months after an intramuscular injection the skeleton and soft tissues still contained 27 per cent of the administered dose.

GROUP VI

Most of the Group VI elements (Table 8) showed some tendency towards association with the circulating red cells (as was noted for the Group V elements). The metal–protein combination becomes more stable with increasing atomic weight of the Group VI metal. Sixteen days after injection only 0.1 per cent of the Se remained in the blood stream, while 10 per cent of the Te was still circulating. After oral administration of Po, the same amount was present in the red cells at 70 days as at 8 to 10 days. The urine was the important route of elimination for Se and Te, and kidney was the only tissue that consistently contained either isotope at higher concentration than the blood. Excretion of Po following absorption from the gastrointestinal tract; i.e. noncolloidal Po, was predominantly fecal, although urinary excretion was higher than was found for intravenously injected colloidal Po. There is some evidence for a greater concentration of Po in tissues such as liver and kidney than can be accounted for by tissue blood content.[58]

The Sub-group VI elements apparently did not combine with the circulating red cells to any significant degree, although radiochromate is frequently the method of choice for *in vitro* red-cell labeling. Urinary elimination of these elements occurred chiefly during the first few hours after injection when the blood level was high. Subsequently, most of the excretion took place via the liver and the gastrointestinal tract. The heaviest member of the group, W, was the most rapidly excreted; 22 per cent of the administered Cr was retained after 16 days, whereas only 2 per cent of the tungstate was still present at 4 days. These elements were initially present in nearly all the tissues. Several days after injection bone, liver and kidney were the only tissues that contained appreciable amounts of Cr and W. No critical organ was determined for Mo.

GROUP VII: THE HALOGENS

The metabolic characteristics of the halogens, Group VII, are shown in Table 9. Excretion tended to be almost exclusively urinary, and soft-tissue concentrations were generally low. The skeletal deposition of F was unique among the members of this group. After an intravenous injection, all of the main group halogens

Table 9. Distribution of radioisotopes of the elements of Group VII, the halogens, in various tissues of the rat after intravenous injection

Radioisotopes	Chemical form administered	Time (hr)	Percentage of absorbed dose					Ref.
			Bone	Liver	Thyroid	GI tract + contents	Excreta	
F^{18}	NaF	9	56.1	<0.1	<0.1	0.8	33.2 *U*	15
Cl	—	—	—	—	—	—	—	—
Br^{82}	KBr	8	—	5.6	—	9.2	14.4 *U*	61
I^{131}	NaI	9	1.2	1.0	23.6	8.4	34.4 *U*	14
At^{211}	NaAt	9	3.7	6.1	1.6	21.8	16.6 *U*	14
Sub-group VII								
Mn^{52}	$MnCl_2$	5	7.1	26.2	<0.1	35.8	—	55
$Tc^{95,96,98}$	(4+ or 6+, hydrated)	24	0.4	0.7	<0.1	9.0	87.3 *U*	7
$Re^{183,184}$	$NaReO_4$	4	1.0	1.1	<0.1	13.2	49.6 *U*	7, 53

were secreted by the gastric mucosa to some extent and were reabsorbed almost completely in the small bowel. This phenomenon shows a definite trend of increasing gastric secretion with increasing atomic number.[60] Fluorine was not concentrated by the thyroid gland, and thus far At is the only halogen other than iodine that has been shown to be accumulated by the thyroid to a significant degree under normal conditions.[14,18] Astatine homologues of the iodinated organic compounds synthesized by the thyroid gland have not been identified.

The metabolic characteristics of Mn, the first member of the seventh sub-group, were quite unlike those of the main halogen group elements and the other sub-group VII elements. Manganese injected intravenously in the 2+ state was accumulated largely by the liver, and rapidly appeared in the gastrointestinal tract. One day after injection the liver still contained 11 per cent of the injected dose, another 11 per cent was found in the gastrointestinal contents, and 41 per cent had been eliminated in the feces. Significant amounts of Mn were found in spleen, pancreas, kidney and skeleton. It was not determined whether the skeletal Mn was deposited in the marrow as colloidal particles or was present in the bone tissue itself in a manner similar to other divalent elements.

There is some evidence for at least transient thyroidal accumulation of the highly hydrated positive valence states of Re and Tc, particularly in rats that have been maintained on a low-iodine diet, or in rats made goiterous with thiouracil.[62] These elements were eliminated almost quantitatively by the kidney and exhibited the gastric secretion and subsequent intestinal reabsorption seen with the elements of the main group.

THE PLATINUM METALS

The distribution of the platinum metals is shown in Table 10. The oxidation states of these elements as they were prepared for injection ranged from 2+ for Pd and Pt to 8+ for Os. Although the trichloride is shown as the injected form of Rh and Ir, it is possible that they existed to some extent as complex chlorides, i.e. $Rh(Cl_6)^{3-}$. Ruthenium, Os, Pd and Pt were administered as complex anions. The subsequent behavior in the circulation and in the tissues would depend on whether they remained in the injected form, or were rendered less soluble (as by hydroxide formation), or were reduced to the metallic state. There seemed to be little tendency for these elements to form radiocolloids following intravenous injection as judged by: (a) the low liver uptakes and spleen concentrations (less than 0.7 per cent/g in all

Table 10. *Distribution of radioisotopes of the transition and platinum metals in various tissues of the rat after intravenous injection*

Radioisotopes	Chemical form administered	Time (days)	Percentage of absorbed dose						Ref.
			Bone	Liver	Kidney	Blood	Muscle	Excreta	
Fe^{59}	$FeCl_3$	3	8.4	20.0	2.1	52.5	7.7	6.3 *F*	44
Ru^{97}	Na_2RuCl_5OH	1	6.5	5.8	2.7	7.4	21.4	19.4 *U*	53
Os^{185}*	$NaHOsO_5$ or OsO_4	1	1.9	3.6	4.5	2.9	3.4	78.7 *U*	64
Co^{60}	$COCl_2$	1	—	6.3	0.8	0.6	—	67.9 *U*	65
Rh^{105}	$RhCl_3$ or $RhCl_6^{3-}$	1	3.7	3.6	2.6	5.9	12.1	47.2 *U*	37
$Ir^{190,192}$	$IrCl_3$ or $IrCl_6^{3-}$	1	3.1	19.3	4.0	6.4	5.6	43.5 *U*	53
Ni	—	—	—	—	—	—	—	—	—
Pd^{103}	Na_2PdCl_4	1	1.0	8.6	8.4	0.8	1.3	74.8 *U*	53
$Pt^{191,193}$	Na_2PtCl_4	1	5.8	12.9	9.9	6.6	11.6	36.6 *UF*	37

* Os injected intramuscularly.

cases), and (b) the similar distribution of Pt after intravenous or intramuscular injection.

While the transition and platinum metals form relatively homogeneous chemical groups, each element possesses a certain degree of individuality, and the biological behavior of carrier-free radioisotopes of these metals agreed with this general picture. Kidney, liver and spleen were the chief deposition sites of the platinum metals, and were the only tissues that consistently showed higher concentrations than the blood. Bone was not a major site of accumulation of any of the platinum metals during the longest period of observation which was 33 days. It has been shown, however, that after prolonged feeding of radioruthenium the bone concentration is built up to potentially hazardous levels, and that skeletally deposited Ru is held more strongly than that in the soft tissues.[66] Except for Pd, the blood levels of these elements were still high 24 hr after injection (compared with many other elements studied), and Ir and Pt were easily measurable in the blood as long as 32 days after injection. Loss of the lighter members of the Group (Ru, Rh and Pd) from the tissues was less rapid than the decline in their blood concentrations, while the decrease in tissue content of Ir and Pt roughly paralleled the decline of the blood concentrations. Retention of Ru and Rh is probably more prolonged than that of the other members of the group. Excretion of Ru was comparatively low, for only 43.3 per cent had been excreted by the seventh day. Initial excretion of Rh was somewhat higher—47 per cent in the first 24 hr—and only an additional 7 per cent was eliminated in the ensuing 5 days. The other members of the group were eliminated much more efficiently: 80 per cent of the Ir in 33 days, 92 per cent of the Pt in 32 days, 89 per cent of the Pd in 7 days, and 79 per cent of the Os in the first 24 hr.

DISCUSSION

The behavior of the elements of a main chemical group is usually more predictable than the behavior of a sub-group element. For example, the alkaline earths are deposited predominantly in bone mineral, while the sub-group elements (Cd and Hg), which are also stable in the 2+ state, are retained tenaciously in the liver and kidney. Less than 2 per cent of either can be found in the skeleton. Relatively uniform behavior was seen in the main groups of the alkali metals, the alkaline earths, the lanthanides and actinides in the 3+ state, the halogens, and the platinum metals. There were some exceptions, particularly among those elements essential to tissue function and survival. In the adult rat, skeletal deposition of Ca was nearly twice that of any other element in the 2+

state, and urinary elimination was one-fifteenth that of the other alkaline earths. Although most of the halogens can be demonstrated to concentrate at least transiently in the thyroid gland, this gland has been found to utilize only iodine. In mammals the ferric–ferrous redox couple is overwhelmingly preferred in metallo-enzyme systems, and the distribution of tracer Fe bore no resemblance to the other transition and platinum metals studied.

Four properties appeared to be the chief determinants of the behavior of a tracer element in the mammalian organism: (a) the oxidation state stable at the pH of the body fluids, (b) the solubility of the stable state, (c) the tendency towards incorporation into organic compounds, and (d) the tendency to be chelated by or to form complexes with proteins in the circulation or in a particular tissue.

Oxidation state was by far the most important factor. Although Tl is a Group III element, the 1+ state is more stable than the 3+ state characteristic of the other members of the group. It was not surprising to find that Tl deposited primarily in the soft tissues, and that its distribution closely resembled the heavy alkali metals.

Almost as important as oxidation state in determining biological behavior was the tendency of many highly positively charged metal ions to assume complex hydrated, oxygenated or halogenated forms. Such complex anions were readily eliminated, particularly by the kidney. For example, radiogermanium (Group IV) was prepared in its most oxidized form and was probably given as the germanate ($NaHGeO_3$) rather than as the tetrachloride. In 4 days 98.5 per cent of the Ge had been eliminated. Sn and Zr which were most likely given as 4+ cations were retained to a high degree and exhibited the distribution typical of bone seekers.

Solubility in the tissue fluids also influenced distribution to a high degree, even in the case of carrier-free radioelements where mass effect need not be considered. Polonium, radiotantalum as Ta_2O_5, and radiogold in any form thus far tested displayed a marked tendency towards radiocolloid formation. They were poorly absorbed after intraperitoneal or intramuscular injection and accumulated rapidly in the reticulo-endothelial tissues after intravenous administration.

Certain of the cations tend to be bound to proteins and present what seem at first to be anomalies. Radiosilver was associated with serum albumen which was then deposited almost quantitatively in the liver and excreted into the intestinal tract. Radioarsenate rapidly entered the circulating red blood cells as an apparently permanent part of the hemoglobin molecule to be eliminated only at the death of the original host cell. This affinity for the protein, globin, was scarcely detectable for the other members of Group V.

Many other radioelements combined with proteins but in a less spectacular fashion. The rare earths in carrier-free form—the actinides—and several other Group III and Group IV elements, formed complexes with the heavier serum proteins with liver proteins, and possibly with the mucoproteins on bony surfaces. In contrast to the majority of the bone-seeking bivalent cations—Hg^{2+}, Cd^{2+} and UO_2^{2+}—apparently combine with some as yet unidentified protein in the renal cortex.

It is possible to make some generalizations for the benefit of the industrial physician and health physicist. With few exceptions, the anions—including most of the halogens and the oxygenated or halogenated oxidation states of the elements of Groups IV, V, VI and the platinum metals—are eliminated quite rapidly, chiefly by the kidney. The monovalent cations are distributed almost uniformly in the soft tissues, and are subsequently excreted by the kidney but with longer turnover times than the anions. The bivalent cations (with previously indicated exceptions) constitute the first major group of bone seekers. They are associated almost exclusively with bone mineral and are eliminated very slowly as the structural elements of the skeleton mature with advancing age. The tripositive ions tend to associate with protein in the circulating blood, in the liver, and quite possibly in bone, and the most important deposition site of each individual 3+ element seems to depend upon the relative stability of the particular protein complex. The fraction laid down in the liver is eliminated quickly, while that deposited in bone is turned

over much more slowly than are the bivalent elements. The quadrivalent cations, such as Zr, Th and Pu, are deposited almost exclusively in the skeleton, and are held there as tenaciously as are the 3+ elements.

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THE NORMAL RADIUM CONTENT AND THE Ra^{226}/Ca RATIO OF VARIOUS FOODS, DRINKING WATER AND DIFFERENT ORGANS AND TISSUES OF THE HUMAN BODY*

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Abstract—New experimental results concerning the normal radium (Ra^{226}) content of food, drinking water, the distribution of normal radium in the human body, and the Ra^{226}/Ca ratio are reported. The daily uptake of radium by a "standard man" amounts to about 3×10^{-12} c, and under normal environmental conditions, 90 per cent of this amount enters the body with food and only 10 per cent with water. The natural radium is not concentrated exclusively in bone. Considerable amounts are distributed throughout the soft tissue and the different organs. Animal experiments (chicken) demonstrated that there exists no dependence on age of the normal radium content. The consequences of these results for the estimation of the distribution pattern of other bone-seekers are discussed.

For about three years the authors have studied the problem of the normal radium content of the human body.[1–6] Recently we conducted extensive measurements of both radium content (Ra^{226}) and the Ra^{226}/Ca ratio of autopsy material and a large number of samples of food and drinking water. For evaluation of the radium content of the ashed and chemically treated samples, we used the "emanation method".

The radon was measured by two types of equipment:

(a) A special vibrating-reed electrometer with two compensated ionization chambers.
(b) An instrument for counting single alpha pulses including an amplifier and pulse-height selector.

Techniques for preparation, measurement of samples and estimations of the possible analytical error have been given in detail elsewhere.[1–7]

RESULTS

The radium content and the Ra^{226}/Ca ratio of different tissues and organs are given in Table 1. The values are averages pertaining to a large number (n) of individual cases. For the most part our samples were obtained from older people, the average age being more than 60 years. None of these persons suffered from metabolic diseases. In nearly all cases death occurred from traffic accidents or heart failure. The specific activity of different tissues relative to bone (tibia) is given in the last column in percentages (tibia = 100 per cent). It is demonstrated that the specific activity of liver, lungs, spleen, kidney and muscle varies between 17 per cent and 46 per cent of the specific bone activity. Radium content and Ra^{226}/Ca ratio for various foods are given in Tables 2, 3 and 4 (Table 2 pertains to food of animal origin and Table 3 to food of plant origin). The natural radium content of different kinds of water is given in Tables 5 (tap water), 6 (river water) and 7 (deep wells and ground water).

DISCUSSION

(a) *Average radium content of the human body*

From Table 1 the average radium concentration in bone is found to be 5×10^{-15} c Ra^{226}/g

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Table 1. *Radium content* (Ra^{226}) *of different tissue samples*

Tissue	*n**	Radium content/g ash ($\times 10^{-14}$ c)	Radium content/g wet tissue ($\times 10^{-15}$ c)	Radium content/g calcium ($\times 10^{-13}$ c)	Radium content/g wet tissue (%)
Bone (tibia, shaft)	56	1.2	5.4	0.3	100
Bone (femur)	37	1.1	4.8	0.5	89
Great gut	25	19.6	0.6	94	11
Small gut	25	15.5	0.7	57	13
Skin	16	17.5	0.7	69	13
Liver	12	17.4	2.0	377	37
Lungs	39	15.0	1.6	84	30
Spleen	32	13.4	1.7	176	32
Muscles	12	24.5	2.5	226	46
Kidney	36	10.2	0.9	59	17
Pancreas	35	14.4	0.8	43	15
Testis	23	2.9	0.3	16	6
Placenta	8	9.9	0.8	24	15
Foetus	9	2.5	0.6	1	11

* The measured samples are averaged from *n* persons.

Table 2. *Radium content* (Ra^{226}) *of various foods*

Food	Starting material (g)	Ca in starting material (g)	Radium content/g starting material ($\times 10^{-15}$ c)	Radium content/g calcium ($\times 10^{-13}$ c)
Pork	1000	0.079	0.8	97
	1000	0.085	1.5	178
Pork liver	300	0.030	0.8	795
Beef	1050	0.096	0.8	88
	880	0.068	0.8	107
Blood (cattle)	2000	0.117	0.2	41
Milk	1000	1.280	0.3	2.1
Milk powder	984	11.700	1.7	1.4
20 eggs	940	0.432	3.1	69
Butter	980	0.538	0.3	5.5
Green herrings	975	3.890	2.8	7
(incl. bones)	985	4.140	4.0	10
Cod (without bones)	600	0.158	4.0	150
Haddock (without bones)	635	0.209	6.3	190
Codliver oil	1000	0.022	4.9	2240

Table 3. Radium content (Ra^{226}) *of various foods*

Food	Starting material (g)	Ca in starting material (g)	Radium content/g starting material ($\times 10^{-15}$ c)	Radium content/g calcium ($\times 10^{-13}$ c)
Bread	1475	0.283	2.6	138
White bread	896	0.840	3.3	34
	1181	0.850	1.7	24
Wheat flour	500	0.102	2.7	136
Margarine	986	0.288	0.1	3.4
Carrots	796	0.260	1.6	49
	787	0.264	1.7	52
	1240	0.460	6.1	229
Cabbage	810	0.758	2.4	26
	823	0.621	1.0	13
Apples	1618	0.085	0.9	150

Table 4. Radium content (Ra^{226}) *of potatoes in different areas of Germany*

Area	Number of samples	Average weight of one sample (g)	Radium content/g fresh weight ($\times 10^{-15}$ c)	Radium content/g calcium ($\times 10^{-13}$ c)
Northern Germany	22	1500	1.0	238
Southern Germany	17	2000	0.6	124

Table 5. Radium (Ra^{226}) *in tap water (drinking water)*

Location	c radium/l. ($\times 10^{-13}$)	Calcium (g/l.)	c radium/g calcium ($\times 10^{-13}$)	c radon/l. ($\times 10^{-10}$)
Frankfurt/M.	2.2	0.040	55	1.3
München	0.9	—	—	—
Northeim	3.4	0.078	44	2.2
Inheiden	2.0	—	—	—
Bochum	0.3	—	—	—
Erlangen	1.5	—	—	—
Badgastein	2.7	0.016	170	—

Table 6. Radium (Ra^{226}) *in river water*

	c radium/l. ($\times 10^{-13}$)	Calcium (g/l.)	c radium/g calcium ($\times 10^{-13}$)
Main	0.7	0.079	8.8
Isar	7.0	—	—
Leine	8.4	0.118	71.0
Regnitz	1.2	—	—

Table 7. Radium (Ra^{226}) *in springs*

Location	c radium/l. ($\times 10^{-13}$)	Calcium (g/l.)	c radium/g calcium ($\times 10^{-13}$)	c radon/l. ($\times 10^{-10}$)
Wells 400–500 m deep				
Hassia-Sprudel Bad Vilbel	91.6	0.272	337	6.5
Friedrich-Karl-Spr. Bad Vilbel	117.0	—	—	—
Kronthal	184.0	0.257	715	10.6
Wells 30–50 m deep				
Erlbach bei Donauwörth	2.6	—	—	—
Bismarckquelle Friedrichsruh	1.3	0.122	10.9	—
Hermannsquelle Mölln	0.7	0.065	10.8	—
Hitzacker	1.4	0.104	13.0	—
Ratzeburg	5.2	0.094	55.6	—

wet bone. Hence the total radium content of the skeleton is estimated at 0.35×10^{-10} c. (Using the data for the "standard man" the weight of the skeleton is assumed to be 10 per cent of a total weight of 70 kg.) The average radium content of the total muscle–tissue complex (43 per cent of the total weight, or 30 kg) is found to be 0.7×10^{-10} c. Skeletal and muscular tissues together amount to 53 per cent of the total body weight and therefore carry 1.1×10^{-10} c Ra^{226}. Assuming for the tissues and organs of the rest of the body (47 per cent) an average specific radium content of about 0.7×10^{-15} c/g wet tissue in accordance with data of Table 1, the activity of this part of the body is about 0.2×10^{-10} c. Hence the normal radium content of the total body is estimated to be about 1.3×10^{-10} c. This value agrees with results given by HURSH and GATES[8,9] (average of 20 persons: 1.2×10^{-10} c)

and STEHNEY and LUCAS[10]. On the other hand the results published by PALMER and QUEEN[11] are considerably lower (average of 50 persons: 0.5×10^{-10} c). In a recent publication WALTON *et al.*[12] have reported radium concentrations in human bone samples from different locations of the world with a world-wide arithmetical average of about 1.2×10^{-14} c Ra/g bone ash. This figure is in good agreement with our values for bone ash given in Table 1 column 3 (1.1 and 1.2×10^{-14} c Ra/g bone ash respectively). The average value of 0.8×10^{-14} c Ra/g ash for the *whole body ash* of 140 cadavers from New York City given by WALTON *et al.* seems to be surprisingly low compared with the concentrations in bone ash alone. The values for cremation ashes reported by HURSH[8,9] and new data concerning the radium content of *whole body-ash samples* measured in Frankfurt (MUTH, STAHLHOFEN and AURAND, In press) give much higher figures in good agreement with our estimations of the whole body radium content from radium concentrations *in bone and in soft tissue*. Differences in ashing procedures may influence the results to a high degree because the fine ash from soft tissue may leave the crematorium immediately through the chimney (MUTH *et al.*,[3] see the summarizing report of SPIERS[13] and SPIERS and BURCH[14].

(b) *Radium intake with food and water*

The radium content of drinking water and well water (Tables 5 and 7) in different areas of Germany is found to be comparable with values given by HURSH[15] and STEHNEY[16] for tap water and deep well water of different regions of the U.S.A. The radium content of food (Tables 2, 3 and 4) varies between 0.1×10^{-15} c and 6×10^{-15} c Ra/g. So far only two papers have been published pertaining to the natural radium content of food: SHANDLEY[17] and BURKSER, SCHAPIRO and BRONSTEIN[18]. Small differences between SHANDLEY's and our results may reflect the variability of material from different geographic locations. This is indicated by our measurements of the radium content of potatoes (Table 4). Samples originating from two different districts of Germany differ in average radium content by nearly a factor of 2. An estimate using the data for the standard man shows that under normal environmental conditions 90 per cent of radium uptake occurs from food and only about 10 per cent from drinking water.

(c) *Natural radium content and total natural alpha-activity*

A few months ago TURNER, RADLEY and MAYNEORD[19–21] reported results of measurements of the total natural alpha-activity of man, animals, food and soil samples. Their values of the total alpha-activity in human bone are about a factor of 10 higher than the Ra^{226} content reported here. Since, according to the findings of TURNER *et al.*, about 40 per cent of the total alpha-activity in the bones of adults is due to radiothorium and its decay products, this factor of 10 is easily explained by the number of alpha-emitting products of the two radioactive series. TURNER obtains tenfold higher values for the total alpha-activity of cattle bone than for human bone. This agrees with our results, that the natural Ra^{226} content of cattle bone is ten times that of human bone. The authors have also measured the total alpha-activity of human bone and tissue samples and have confirmed the data given by TURNER *et al.* These results will be published elsewhere.[22]

(d) *Dependence of radium content on age*

During the past year the authors have studied the variation of the natural radium burden of chickens with age. It is found that at the end of the period of bone growth, i.e. after about 60–100 days, the specific bone activity remained constant, and compared with that of human beings (Fig. 1). There exists no increasing

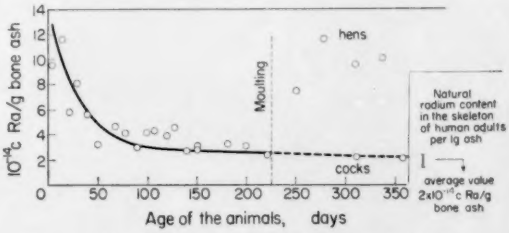

FIG. 1. Dependence on age of the natural radium content in the skeleton of chickens (activity/ 1 gm bone ash).

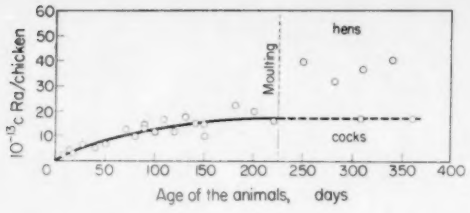

FIG. 2. Dependence on age of the natural radium content in the skeleton of chickens.

radium content with age (RAJEWSKY *et al.*[23] and HANTKE[7]). Fig. 2 gives the corresponding dependence on age of the natural radium content of the whole skeleton of chickens. After moulting the hens differ from the cocks, the hens having a higher radium content than the cocks which show no moulting.

Measurements of the radium content of human bone samples of different age-periods confirmed the results with animals: there is no higher radium content with increasing age. The results of these experiments are given in detail and discussed by HANTKE[7].

(e) *General conclusions concerning the continuous daily intake of bone-seekers*

The normal radium-content data given in Table 1 demonstrate that the specific activity of special organs and tissues under conditions of continuous daily uptake is comparable with specific activity of bone. This is in contrast to cases of chronic radium poisoning, where the total radium deposit occurs only in bone. The Ra^{226}/Ca ratio in bone amounts to 0.4–0.7, in muscle tissue to more than 200, and for other organs is between 16 and 380 units of 10^{-13} c Ra^{226}/g of Ca. In food the normal Ra^{226}/Ca ratio varies between 2.1 for milk and more than 700 for pork liver. Hence the Ra^{226}/Ca ratio in human soft tissues compares with that of food, while in bone there exists a strong discrimination against radium as compared with calcium. The data in Table 1 demonstrate that in contrast to cases of chronic radium poisoning the continuous intake of small amounts of radium necessitates consideration of the radiation exposure of other organs (perhaps including gonads) as well as bone. It should be pointed out that this result may be important for the problem of continuous uptake of small amounts of Sr^{90}. So far Sr^{90} content has been measured only in bone (KULP *et al.*[24–27]) and only the radiation exposure of bone has been discussed. It seems desirable that future investigations study the Sr^{90} content of different organs and tissues, particularly the gonads. Only such data can provide a good basis for future general discussions of the Sr^{90} problem. This is especially important in view of the increasing Sr^{90}-level in mankind with continued atomic bomb tests.

Fig. 3 gives an example of the distribution pattern of natural radium content in bone (74 different samples). The curve is obviously not of the Gaussian type. LIBBY[28] and KULP *et al.*[24–27] discussing the effects of incorporated Sr^{90} due to test explosions, assume a Gaussian distribution. LIBBY attempts to show that the distribution of the natural nonradioactive Sr is a normal one too, and that therefore the probability of exceeding the threefold average is 5×10^{-8}. However even in his presentation, summarizing a total of 297 cases, already several cases exist above the threefold average value. LIBBY also assumes, based on results from PALMER and QUEEN[11], a normal Gaussian distribution for the normal radium content. Our results in Fig. 3 indicate that this assumption may not be justified. A more exact statistical analysis of this problem (AURAND[29]) is in preparation.

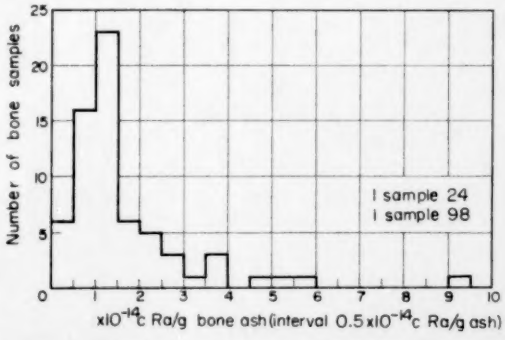

FIG. 3. Distribution pattern of Ra^{226} in human bone.

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THE PREPARATION OF THIN, UNIFORM, RADIOACTIVE SOURCES BY SURFACE ADSORPTION AND ELECTRODEPOSITION*

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Abstract—Thin, uniform, radioactive sources were deposited by several methods on thin films composed of a sandwich of metal, polystyrene and formvar. Sources of cerium-144, yttrium-91, scandium-46 and ruthenium-106 were prepared by adsorbing the hydrolyzed radionuclides on gold-coated films, while selenium-75, ruthenium-106, iridium-192, cobalt-60 and chromium-51 were adsorbed in the elemental form on magnesium. Ruthenium-106, silver-110, mercury-203, cobalt-60, indium-114, iron-59, chromium-51 and gold-198 were electrodeposited on gold, the plating potential being supplied either externally or by a dissolving magnesium cathode. The uniformity of the sources was evaluated by radioautography, and in the case of Au^{199} by examining the conversion electron spectrum with a homogeneous-field solenoidal-type beta spectrometer.

INTRODUCTION

A SOURCE for β-spectrometry should be uniformly thin and should be deposited on a thin backing in order to reduce self-absorption and back-scattering of β-particles to a minimum.[1,2] In addition, it is desirable to have as high a source activity as possible, and to deposit the radioactive material in a definite geometrical pattern. Information on source preparation other than by solvent evaporation is summarized in Table 1.

The three most common source-preparation methods are solvent evaporation, vacuum thermal evaporation and electrodeposition.[74,79,84,85] Sources are deposited most conveniently and completely by the first method, in which a drop of the radioactive solution is pipetted on to the source backing and is then heated to drive off the solvent. During evaporation, crystals containing the radionuclide are deposited at the periphery of the liquid, resulting in a nonuniform source. Somewhat greater uniformity is obtained by reducing the surface tension of the solvent, for example with insulin[26] or tetraethylene glycol.[79]

Most radionuclides can be deposited uniformly with vacuum thermal evaporation[43] from a hot filament. However, there must be an appreciable distance between the filament and the organic film used as source backing to prevent the destruction of the latter by heat radiated from the filament; hence a large fraction of the radionuclide will usually be lost on the walls of the evaporator. This loss of activity may be eliminated either by preparing a relatively volatile salt of the radionuclide and heating it in a chamber pierced by a pinhole, which allows the radioactive material to escape to the film;[9] or by depositing a non-radioactive source on a backing at a distance from the filament, and then producing the radionuclide by an appropriate irradiation.

* A preliminary account of this work appears in *Bull. Amer. Phys. Soc.* Ser. II, **2**, No. 5, 279 (1957). A more complete description is available as ORNL-2419, 25 November 1957.

This work has been submitted by one of us (R. L. B.) in partial fulfillment of the requirements for the Master of Science degree at Vanderbilt University.

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§ Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

Table 1. *Methods of source preparation**

Element	E	V	A	M	R	D	Element	E	V	A	M	R	D
H						3	La				33		
C						4	Hf		50		13		
Na			5				Re	51, 52	52				
P†	6	7	8				Ir†		53				
S		9					Au†	54	43			55	
K		10, 11					Hg†		41		13		
Sc†		12					Tl†	56			49	57	
Ti				13			Pb	58, 59	60	61	13	59	
Cr†							Bi	58, 62	63	61		59	
Fe†	14, 15							44, 59					
Co†	16, 17	9	19				Po	64, 65		59		59	
	15, 18							59					
Ni	20	9					At	66	66	66, 67			
Cu	21	22, 23	24				Rn					68	
Zn	25, 15	26	27				Ra	69					
As		28		13			Ac	64, 70					
Se†								69					
Br				13			Ce†				33		
Kr				13, 29			Pm	72	71, 4		33		
Rb†		30, 31		13			Sm				73		
Sr†	32	7		33			Eu		53		73		
Y†		7		33			Er				49		
Mo				13			Tm				49		
Ru†	34, 35			33			Yb				49		
Rh	36						Th	70, 74		61			
Ag†	36, 37	38	27, 39					75, 69					
			40				Pa	76, 77		78			
Cd	36	41					U	64, 74	74, 79				
In†	42							79, 75					
Sn	36, 43						Np	64, 74					
	44							75, 80					
Sb	36, 44	45					Pu	80, 64	74, 79				
Te	36							74, 79	82				
I			8	13				75, 81					
Xe				46, 13		47	Am	**75**	82				
Cs		43		49, 13		48	Cm	75, 83	82				
Ba			8	33									

* Numbers correspond to the references at the end of the paper; the letters indicate: E, electrodeposition; V, thermal volatilization; M, electromagnetic separation; D, deposition from gas phase; A, absorption from liquid phase; R, recoil separation.

† Indicates nuclides studied in this research.

Electrodeposition of radionuclides has been used most frequently in order to prepare α-sources,[74,79,85] make chemical separations,[36,44,85] or study the behavior of ions in very low concentrations.[87] The preparation of spectrometer sources differs only in that the cathode is a thin metal foil or a metal-coated plastic film. Conditions for depositing massive amounts generally apply to the deposition of radionuclides; however, the Nernst equation—which is used to calculate the e.m.f. needed for electroplating—does not in all cases describe the

latter.[88-90] Moreover, the current is carried almost exclusively by ions other than those of the radionuclide which is usually present in a comparatively small concentration. Thus both voltage and current must usually be obtained by trial and error.

Other methods of source deposition utilize separation of the radionuclide from its non-radioactive isotopes with a mass spectrograph, the target of which ultimately serves as the source backing;[13,29] deposition of gases and their decay products on metal cathodes;[47,59] and the collection of radioactive atoms recoiling after formation from neutron[55] or heavy ion bombardment,[68] or from α- or β-emission.[57,59]

Two other preparative methods—surface adsorption and spontaneous deposition of ions from electric cells—should find general application, and have been utilized in this work. Surface adsorption is used here rather loosely to describe possible mechanisms such as ion exchange, the formation of chemical compounds, ionic interchange between a substance in solution and one on the surface, and deposition because of the existence of a local difference in potential. While some adsorption of radionuclides on surfaces occurs under almost all conditions,[5,8,61] circumstances most favorable to adsorption are probably those under which radiocolloids[39,91] are formed. Adsorption of radiocolloids has been employed in this work for a number of radionuclides either by hydrolyzing them or by reducing them to the insoluble elemental form. An example of the use of the latter technique can be found in the separation of the more noble elements of the natural radioactive chains (such as polonium) from the others by reduction with, and adsorption on, plates of a less noble metal (such as silver or nickel).[59,87] Deposition from an electric cell is achieved by the same reduction process, but the radionuclide is deposited on an inert anode rather than on the reducing metal.

In addition to using these source-preparation methods, radionuclides were deposited for purposes of comparison by solvent evaporation and by electrodeposition with an external source of e.m.f. Uniformity of the sources was compared by radioautography, and an electrodeposited source of gold-199 was examined with a β-spectrometer to detect the results of self-absorption or back-scattering in the widening and lack of symmetry of the conversion electron lines.

PROCEDURE

All radionuclides were obtained from the Isotopes Division of the Oak Ridge National Laboratory in tracer amounts and were diluted to a concentration of approximately 10^{-2} μc/ml in 0.1 M acid solutions (mercury and silver were in nitric acid, all others in hydrochloric). The concentration of the radioisotopes, calculated from the given specific activity, was less than 10^{-6} M for mercury-203, silver-110, iridium-192, chromium-51, indium-114 and iron-59; less than 10^{-7} M for scandium-46, selenium-75 and cobalt-60; and less than 10^{-8} M for the remainder.

Sources were deposited on a film consisting of 15 μg/cm^2 of formvar to provide strength, covered by 15 μg/cm^2 of polystyrene to resist chemical action. The films were prepared by dipping a glass plate successively in 0.5% formvar in ethylene dichloride solution and 1% polystyrene in benzene. The glass plate was then immersed in water to permit the films to float off on the water surface.[92] A metal film, 30 μg/cm^2 thick, of gold or magnesium was deposited on the films by vacuum thermal evaporation.[93] The metal film was used both as an electrode in depositing the source, and as a conductor for grounding the source in the β-spectrometer. Sources prepared especially for the β-spectrometer[46] in the shape of an annulus 13 mm in diameter and 0.6 mm wide were deposited on the films by masking the residual area. A complete discussion on the preparation of thin films may be found in a report edited by HUDSWELL and MANDELBERG[85].

The sources were deposited in a container consisting of the source backing—the films glued to metal rings—clamped to a glass cylinder, and made watertight with a rubber washer. The glass was coated with "Desicote", a silicone, to prevent the adsorption of the radionuclides on the glass.

The type of stirring rod and the metallic coating of the film depended on the method of source deposition. These methods were:

(1) Adsorption after hydrolysis: glass rod ("Desicote" covered); gold metal.
(2) Adsorption after reduction: glass rod; magnesium metal.
(3) Spontaneous electrodeposition: magnesium rod; gold metal.
(4) Electrodeposition with external e.m.f.: platinum rod; gold metal.

For method (1) 15 M ammonium hydroxide was added to neutralize 10-ml volumes of acid solution, while for methods (2) and (3), the 0.1 M acid solutions were used unaltered. Electrodeposition with an external e.m.f. (method (4)) was performed on the stock solutions except when almost no deposition occurred, as in the case of iron and cobalt. In order to electrodeposit these more effectively, iron was then reduced to a valence of 2 with hydrogen sulfide, and cobalt[17] was converted to an ammonia complex by refluxing with ammonium peroxydisulfate and neutralizing with ammonium hydroxide.

The electrical circuit was completed for spontaneous electrodeposition by connecting the stirring rod and metallic film coating with a wire. For electrodeposition with an external e.m.f., a battery, rheostat, voltmeter, ammeter and switch were inserted between the stirring rod and film coating. The magnitude of the applied potential was approximately 4 V, which was sufficient to obtain rapid deposition; the current, determined by the concentration of the acid, was approximately 40 mA.

The fraction of the total activity deposited on the films was found by counting aliquots of the supernatant solution before, during and after deposition, with a well-type NaI(Tl) γ-detector or an end-window GM counter. Uniformity of the sources was compared by making radioautographs on DuPont-502 films. The film was exposed to the sources for approximately 1 hr (the exact time for obtaining satisfactory contrast depending on energy and intensity of β-radiation), placed in DuPont X-ray developer for 3 min, rinsed, fixed in X-ray hypo for 20 min, and rinsed again. The variation in film darkening was measured with an Ansco densitometer, and a value of relative uniformity was obtained by dividing the extreme values of twenty readings by the average of these readings. The uniformity of the gold-199 source was estimated from the variation of conversion electron peak width with energy, and the deviation from symmetry of the peaks, as observed in the β-spectrometer.[46]

RESULTS AND DISCUSSION

To compare the uniformity of sources deposited by the four methods listed and with sources prepared by solvent-evaporation, the radioautographs of ruthenium-106 shown in Fig. 1 were taken. The first solvent-evaporation source was prepared by pipetting 0.025 ml of ruthenium solution onto the film and then heating; the second, by adding 0.025 ml of insulin (40 units/ml) before drying the source. Fig. 1(a) and (b) indicate that insulin prevented deposition of all the radioactivity in concentric circles without, however, achieving complete uniformity. The relative uniformity of solvent evaporation sources prepared without and with insulin was 1.84 and 1.64, respectively.

All of the tracer radionuclides in 0.1 M acid solution were brought into contact with gold-covered films to determine their adsorption in the absence of radiocolloid formation or electrodeposition. All were adsorbed to some degree, the adsorption ranging from less than 1 per cent of the total activity for cerium-144, yttrium-91, scandium-46, cobalt-60, chromium-51, selenium-75, ruthenium-106 and iridium-192, to 87 per cent of silver-110 and 99.9 per cent of mercury-203. The small but finite uptake on surfaces of radionuclides from solutions in which they are ionic has been described previously,[5,8,19,61] and has been found to be proportional to the concentration of the radionuclides;[61] the much more complete uptake of certain metallic ions has been attributed either to a chemical reaction with the metal surface, or to electrodeposition caused by the formation of a local cell by impurities on the surface.[8,19]

By neutralizing the acid solution, the adsorption of cerium-144, yttrium-91, scandium-46, and ruthenium-106 was increased to between 10 per cent and 30 per cent, the exact fractional deposition varying appreciably within these limits with pH, and even in duplicate samples at the same pH. The rate of adsorption is relatively great for the first hour and becomes small thereafter, as indicated by a representative uptake curve for cerium-144 in Fig. 2; the uniformity of adsorption, with a relative value for the radioautograph in Fig. 1(c) of 1.18, is greater than that by solvent evaporation, but less than that for sources prepared by any of the other methods described. The greater source uniformity achieved by depositing an insoluble compound instead of evaporating the solvent has been reported.[94] An analogous situation exists in the deposition of radionuclides in such small concentrations that radiocolloidal behavior rather than solubility is involved. Both the irregular adsorption and the dependence of adsorption on the presence of other ions is typical of radiocolloidal behavior.[39]

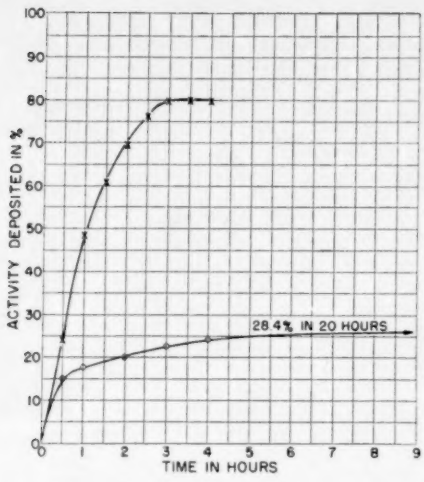

FIG. 2. Adsorption of Se^{75} on magnesium (crosses) from 0.1 M HCl and of Ce^{144} on gold (circles) from 0.1 M NH_4Cl.

By substituting for the gold coating on the film a metallic coating much less noble, such as magnesium, the deposition of selenium-75, ruthenium-106, iridium-192, cobalt-60 and chromium-51 from 0.1 M acid increased to 79, 72, 69, 67 and 34 per cent, respectively. As indicated by a typical uptake curve for selenium-75 in Fig. 2, maximum deposition was attained in approximately 3 hr. After another 2 hr, the radionuclides were again found in solution because the metallic film had dissolved in the acid. The relatively high fractional adsorption is probably caused by the reduction of the radionuclide to the elemental form in the vicinity of the magnesium, rather than throughout the solution. The uniformity of the source—1.02 for the radioautograph in Fig. 1(d)—may be ascribed to the continual exposure of fresh adsorption sites as the magnesium dissolves. Sources deposited in this manner were the most uniform of all studied and, as indicated here, can be prepared for many metallic radionuclides, rather than only for the most noble ones. The disadvantage of the method lies in the continual dissolving of the thin magnesium coating, which prevents more complete deposition of the radionuclides.

To overcome this limitation, a conventional electric cell was prepared by inserting in the acid solution of the radionuclides a magnesium cathode connected externally to the gold-coated film. Thus, while the mechanism of reducing the radionuclide with the magnesium remains the same, the radionuclide now presumably travels to the gold anode as an ion, and is deposited there as the element. The radionuclides listed in Table 2 were readily deposited

Table 2. Electrodeposition of radionuclides

Radionuclide	Chemical form	Solution	Externally applied e.m.f.				Spontaneous deposition (magnesium cathode)	
			e.m.f. (V)	Current (mA)	Time (hr)	Fraction deposited (%)	Time (hr)	Fraction deposited (%)
Ruthenium-106	$RuCl_3$	0.1 M HCl	3.8	55	3.5	62	8.0	96
Silver-110	$AgNO_3$	0.1 M HNO_3	2.8	20	3.0	92	5.0	98.7
Mercury-203	$Hg(NO_3)_2$	0.1 M HNO_3	3.0	20	3.0	94	8.0	97
Cobalt-60	$CoCl_2$	0.1 M HCl	2.9	55	4.0	1	6.0	97
	NH_3 complex		4.2	14	4.0	91		
Indium-114	InCl	0.1 M HCl	3.4	50	3.0	33	3.0	98.6
Chromium-51	$CrCl_3$	0.1 M HCl	2.8	40	4.5	15	5.0	97
Gold-198	$AuCl_3$	0.1 M HCl–HNO_3	4.0	40	4.0	95	5.5	98
Iron-59	$FeCl_3$	0.1 M HCl	3.2	55	3.0	2	6.0	99.2
	$FeCl_2$		5.0	40	5.0	95		

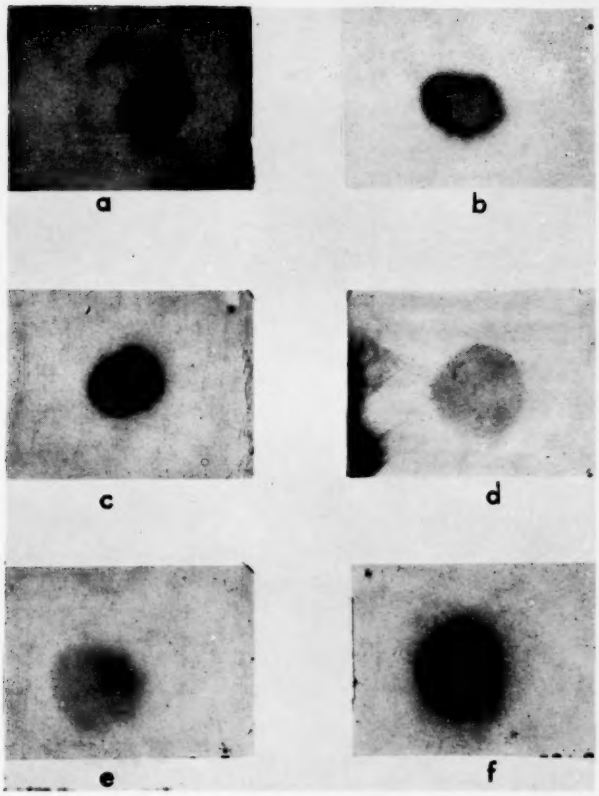

FIG. 1. Radioautographs of ruthenium-106 sources prepared by: (a) solvent evaporated on gold film; (b) solvent evaporated with insulin on gold film; (c) radiocolloidal adsorption on gold film; (d) radiocolloidal adsorption on magnesium film; (e) spontaneous electrodeposition on gold film; (f) electrodeposition with applied e.m.f. on gold film.

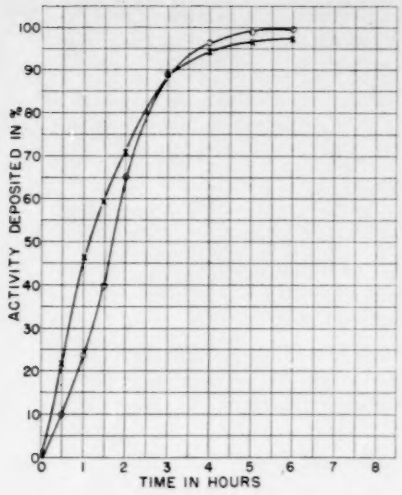

FIG. 3. Electrodeposition of Fe^{59}. Results with spontaneous deposition (see text) are shown as circles. Data obtained by electrodeposition with an e.m.f. of 5 V and a current of 55 mA are indicated by crosses.

by this method, with a fractional deposition exceeding 96 per cent in all cases. The relative uniformity of the radioautograph in Fig. 1(e) is 1.04; a typical uptake curve for spontaneous electrodeposition is given for iron-59 in Fig. 3.

For comparison, all of the radionuclides listed in Table 2 were subjected to electrodeposition with an external source of e.m.f. The gold film thus became the cathode of the plating cell, and the platinum stirring rod the anode. Surprisingly, while initial deposition with a sufficiently high external potential (from 2.8 to 5.0 V) exceeded fractional deposition by spontaneous electrodeposition, the ultimate uptake was not as complete, as shown in Fig. 3. An additional advantage of spontaneous electrodeposition was demonstrated for iron and cobalt, both of which were deposited spontaneously from the original solution, but required chemical treatment prior to electrodeposition with an external e.m.f. While this does not mean that the chemical state of the radionuclide exerts less influence in spontaneous electrodeposition,[35] source preparation by this method may be more convenient for a number of radionuclides. The relative uniformity of a source deposited with an external e.m.f. (Fig. 1(f)) was 1.05, differing little from spontaneous electrodeposition.

Sources prepared by radiocolloidal adsorption and by spontaneous electrodeposition were as adherent as sources prepared by electrodeposition with an external e.m.f., and more adherent than sources prepared by solvent evaporation. From 10 to 20 per cent of the latter, but only 5 to 10 per cent of the former could be removed by rubbing the source with tissue paper.

To determine if the methods used with radioactive tracers were applicable to the preparation of β-spectrometer sources where the activity required was much higher, a source of Au^{199} was prepared.[95] The radionuclide was produced by the irradiation of 1 g of platinum for 60 hr in the Oak Ridge reactor, and by then allowing the platinum-199 to β-decay to gold-199. The platinum metal was dissolved in aqua regia and evaporated to dryness. The nitrate was destroyed by evaporating with hydrochloric acid, and the platinum and gold were taken up in 6 M hydrochloric acid. The gold-199 was extracted into ethyl acetate from the platinum. The gold solution was washed twice with 6 M hydrochloric acid and then dried. The gold was dissolved in 0.5 M aqua regia, and the source deposited from that solution. The γ-spectrum of the final solution indicated that 99.3 per cent of the activity was gold-199, and the remainder gold-198, suggesting that the platinum contained a small amount of gold metal. Assuming the gold-198 to result from the neutron irradiation of gold metal, 21 μg of stable gold as calculated to be in the gold-199 solution. The presence of some stable gold in the solution was fortunate in one respect, since in hundredfold dilutions of the gold solution the radioactivity was adsorbed readily on glass containers even from strong solutions of aqua regia. Thus, in order to count diluted solutions of the gold in aqua regia, it was found necessary to add gold chloride carrier.

The β-spectrometer source was prepared by electrodepositing a portion of the gold-199 with an externally applied e.m.f. of 4 V at a current of 30 mA. Electrodeposition was preferred to adsorption in order to deposit the gold on a gold-coated ring, rather than on the whole film

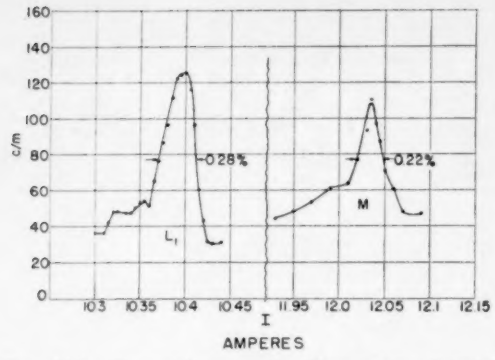

FIG. 4(a). Internal conversion line spectra from the 50 keV γ-ray from Hg^{199}. Lines L_2 and N, O and P were not found. K line lay below the transmission cut-off of the spectrometer.

surface; spontaneous deposition was not used because a short deposition time was required to prevent dissolving the gold ring (since the aqua regia which maintained the radioactive gold in solution equally readily dissolved the gold coating on the film). In the 30 min available, 20 per cent of the radioactive gold was deposited.

Duplicate sources were prepared and examined in the spectrometer to obtain the mercury-199 conversion electron peaks[96-98] given in Fig. 4, and the relative areas under the peaks listed in Table 3. The ordinate is the observed counts per minute and the abscissa is the magnet current which is proportional to the momentum of the electrons which are being observed. The energies corresponding to these momenta may be obtained from Table 3; i.e. the L_I line from the 50 keV γ-ray lies at 10.4 A which is an energy of 35.1 keV, etc. The widths of the peaks are measures of the source thickness in that any absorption of β-ray energy by source material would produce line profiles which would increase in width as lower and lower energy peaks were examined. The spectrometer operates with an inherent line width of 0.25 per cent and this was observed within experimental error for all peaks consisting of single lines. The K and L peaks had a width at half height of 0.25 ± 0.03 per cent. The M and N lines were somewhat wider owing to the fact that each represented the superposition of several closely spaced lines. Corrections were made for the transmission losses of all conversion electrons except those of 50 keV, since the transmission in that energy range decreases very sharply owing to the counter window thickness. Based on a total gold content of 7 μg of solution in the cell and 20 per cent deposition on a ring surface area of 0.3 cm^2, the minimum source thickness is 5 $\mu g/cm^2$. The actual source thickness, judging from the symmetry of the peaks and the lack of

Table 3. *Conversion electrons from* Hg^{199}

Conversion	Relative intensity	Conv. line energy (keV)	γ-Energy (keV)	Average γ-energy (keV)	Previous results
L_I		35.1	50.0	49.6	50.6[97]
M		46.1	49.2		49.8[96]
					50.0[98]
K	100	74.2	158.0		
L_I	15	143.26	158.1		
L_{II}	79	143.90	158.1	158.1	157.5[97]
L_{III}	54	145.80	158.1		158.5[96]
M	31	155.50	158.3		158.5[98]
N	10	157.80	158.1		
K	80	125.6	208.7		208.1[97]
L_I	13	193.5	208.4		208.3[96]
M	2	204.0	207.1	207.8	208.5[98]
N	1	206.8	207.1		

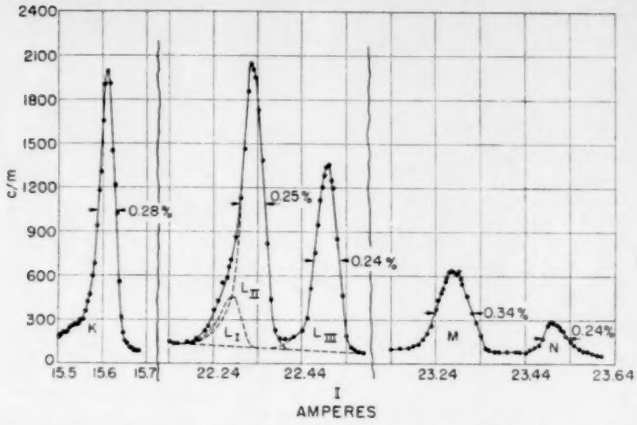

FIG. 4(b). Internal conversion line spectra from the 158 keV γ-ray from Hg^{199}.

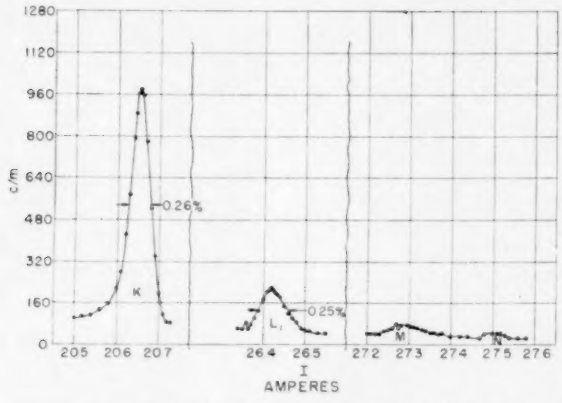

FIG. 4(c). Internal conversion line spectra from the 208 keV γ-ray from Hg^{199}.

appreciable peak widening at low energies, is probably only slightly greater. The maximum thickness that can be calculated by using the LANDAU straggling theory[99] is 50 $\mu g/cm^2$, assuming that the 0.03 per cent variation in peak width is not statistical but rather the result of a thick source.

The ratios of the lines shown in Fig. 4(b) which arise from conversion of the 158 keV γ-ray were corrected for spectrometer transmission and compared with conversion coefficients calculated by M. E. ROSE* for $E2$ radiation. The experimental values for the ratios $K/L_I/L_{II}/L_{III}/M/N$ were 1/0.15/0.79/0.54/0.31/0.10 as compared to the theoretical values for $K/L_I/L_{II}/L_{III}$ of 1/0.13/0.80/0.61. In Fig. 4(c) the ratios of the lines $K/L_I/M/N$ arising from the 209 keV γ-ray were found to be 1/0.16/0.03/0.01 as compared to values for $M1$ radiation for $K/L_I/L_{II}/L_{III}$ from ROSE's tables of 1/0.14/0.013/0.0012.

* The writers are indebted to Dr. ROSE for making available to them the tabulation of coefficients in advance of publication.

CONCLUSION

Thin, radioactive sources at tracer concentrations were prepared on thin, metal-coated polystyrene–formvar films by radiocolloidal surface adsorption and by electrodeposition. Adsorption was more complete and more

uniform by reducing metallic radionuclides to the element than by hydrolyzing radionuclides; in electrodeposition, the spontaneous method was slightly more complete, and in some cases more simple, than the method of utilizing an external e.m.f. Sources by all methods studied were more uniform than sources prepared by solvent evaporation. A high specific activity source of gold-199, prepared by electrodeposition, gave conversion electron distributions in a β-spectrometer expected from the spectrometer design and not unduly widened by source self-absorption or backscattering.

The preparation of uniform sources by reduction and spontaneous electrodeposition should be applicable to many radioactive metals (and possibly metal oxides)[69] by choice of suitable reducing metal. Most other radionuclides can be deposited as radiocolloids, not necessarily as hydroxides, but possibly in other insoluble forms. The chemical form of the radionuclide, its concentration, and the kind and concentration of other substances in solution affects the deposition of the source, and may prevent it. However, the methods described are simple, generally applicable, and should be considered as alternatives to such deposition methods as vacuum thermal evaporation.

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HEALTH PHYSICS FIELD OPERATIONS IN ATOMIC ENERGY COMMISSION WEAPONS-TESTING PROGRAM

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Abstract—Health physics in the support of field operations at the Nevada Test Site and Eniwetok Proving Ground is briefly discussed. At the test sites, the terms "radiological safety" and "health physics" are used interchangeably. Radiological safety requirements necessary to control radiation exposure of all participating personnel at both sites are presented. The organizational setup for health physics support services and assignment of responsibilities for control of radiation exposure to personnel are discussed. The necessary supporting rad-safety functions such as training of monitors, use of aircraft and instrumentation, are included.

1. INTRODUCTION

It is generally believed that participation in weapons testing is an exciting and glamorous business. However, after participation in one or two detonations, the health physics aspects of weapons testing becomes a routine task which requires long duty hours, work under adverse weather conditions, and the ability of health physics personnel to adapt themselves to a frequently changing operational condition.

Health physics in field operations deals specifically with the protection of personnel from the hazards of radiation through control of contaminated areas, materials, vehicles and other equipment. Complete and thorough indoctrination of all personnel in the reasons for radiological safety measures is essential in order to maintain adequate control in the field. The organization responsible for the health of personnel in the field is advisory in nature. However, violation of rad-safety regulations is considered by the test organization to be a serious offense. Through administrative channels, depending upon the seriousness of the offense, an individual can be denied access to the test site, or reported to his home organization so that proper action is instigated to prevent recurrence. Co-operation of the field workers in complying with the standard operating procedures is of tremendous importance in keeping the dosage of workers below the maximum permissible exposure. The maximum permissible dosage for test personnel established by current directives is 3 rems per quarter with the further limitation of no more than 5 rems per year. The m.p.e. is determined by Headquarters, Atomic Energy Commission and sent to the Test Manager or Task Force Commander as a directive.

2. RESPONSIBILITY AND ORGANIZATION]]

The Atomic Energy Commission is responsible for assuring the protection of the health of personnel and the general public with respect to operations in the Commission's programs. The Office of Test Operations, Albuquerque Operations Office, Atomic Energy Commission has contracted to Reynolds Electrical and Engineering Company, Inc., the on-site radiological safety support services for the Nevada Test Site. At the Eniwetok Proving Grounds Holmes and Narver, Inc., Engineers–Contractors, is the contract organization for the radiological safety between full-scale tests. During the test periods Joint Task Force Seven is responsible for the protection of the health of all participating personnel. The Task Group Commanders are in turn responsible for the radiological safety of their own personnel. The radiological safety unit of Task Group 7.1 is responsible for providing the services and conducting the health physics operations during test periods at Eniwetok Proving Grounds. The radiological safety organization of Task Group

7.5, Holmes and Narver, Inc., assists Task Group 7.1 in providing this service. Full-scale tests have in the past been conducted at Nevada Test Site during the odd years and at Eniwetok Proving Ground during the even years. The organization necessary for support of these programs is usually increased from approximately fifteen during the interim period to one hundred during the peak of the operation. These figures are applicable for both test sites.

3. TRAINING

The number of participants involved in test operations increases rapidly at the beginning and then declines as the series nears an end. An efficient training group within the contractor's radiological safety organization is essential, particularly during the buildup phase of an operation. This group is responsible for training all new health physics employees for a variety of jobs within their own organization. In addition, personnel selected to be project monitors require detailed instruction in the operational procedures and in their responsibilities for carrying out these functions. A 3-day basic project monitor's course covering basic radiation subjects and monitoring techniques is given to train the project personnel to act as monitors on a recovery mission. The course is repeated at frequent intervals until all personnel desiring the course have received the training. A refresher course is always necessary, for many of the project personnel have had previous experience but need to be brought up-to-date on the rad-safety procedures which will be used in the operation. These courses are conducted at the Test Site to save in time and money, and also to provide realistic field training. The employees responsible for health physics during the interim period are the nucleus upon which the test radiological safety organization is built.

4. OPERATIONAL CONCEPTS ENIWETOK PROVING GROUND

The initial radiological survey and subsequent resurveys are performed at Eniwetok Proving Ground by means of helicopter (Model H-19). This type of aircraft is ideally suited for Pacific operations where the only other means of travel is by water. Traveling by small boat to the shot island, making a vehicle ground survey, and returning by small boat is much too time-consuming. The information obtained in this manner comes too late to be utilized to the maximum extent. At Eniwetok Proving Ground the Task Group Commander is interested in what the radiation reading is at a particular point or area, and is not particularly interested in producing a detailed isodose map of the atoll. The majority of the scientific recovery missions are made by helicopter to a specific station or area. The recovery personnel make a rapid entry into the contaminated area, obtain their data or equipment, and return to the base island as rapidly as possible. As a result of these considerations it is much more feasible to conduct the radiological surveys by helicopter. An hour's flight is usually sufficient to perform a radiological survey of the shot islands. A Jordon survey instrument which has a logarithmic scale reading a maximum of 1000 roentgens per hour (r/hr) is used. The ion chamber, previously calibrated against a cobalt-60 source, is located on the step of the helicopter. The readings taken by the aerial survey teams are radioed back, via the aircraft control net, to the Rad-safe Center where they are plotted on a suitable map of the atoll. This map is used as a reference for briefing all personnel entering the contaminated island or area.

The health physics controls at the Eniwetok Proving Grounds are much easier than at the Nevada Test Site since all transportation is rigidly controlled by the various operations sections of Joint Task Force Seven. Check stations established at the air-strip and at the boat landing are sufficient to control the entrance of all personnel. Even these controls will not prevent overexposures of project personnel if they become too engrossed in their recovery operations and fail to accept the recommendation of the project monitor who is assigned to each party to insure compliance with the health physics rules and regulations.

Resurveys of the shot areas are made on a continuing basis as long as the contaminated shot island is of interest to the participating personnel. In many cases special surveys have to be made to determine the radiation level at specific locations so that recovery or construction operations can be properly planned.

5. OPERATIONAL CONCEPTS NEVADA TEST SITE

With each succeeding test series, new methods of device placement have been used in an attempt to reduce fallout off-site. This has, of course, changed the expected dose rate contours and the amount of fallout falling on the test site. During previous test series, the majority of shots were air drops or on towers. To minimize off-site fallout, the heights of these towers increased as follows: 100 ft, 300 ft, 400 ft and 500 ft; and, in the last series, one 700 ft tower was used. A point of diminishing returns has about been reached. As towers become higher the amount of material in the tower eventually reaches the point where the amount of material is nearly as great as the dirt that would be sucked up if the fireball touches the ground. Two new methods of weapons placement used during the 1957 test series in Nevada were the use of captive balloons to "hang" the device on, and the placing of the device underground to insure maximum containment of all radioactive material.

Comparison of a tower and a balloon shot is of interest to show the "savings" in money, time and radiation exposure inherent in a balloon detonation. Table 1 compares a balloon and a tower shot. In this table *D*-day signifies shot day.

Figs. 1 and 2 show typical on-site fallout dose-rate contours for a balloon and a tower shot. Most of the radioactivity from the balloon shot is from neutron activation in comparison to the tower shot where the activity is from fission products.

Table 1. *Comparison of tower and balloon detonation*

	Tower	Balloon
Burst height (ft)	500	1500
Yield	½ nominal	½ nominal
Delays due to weather conditions	many	few
Entrance into the shot area	delayed	immediate
Area access	general area	limited area
Recovery (D and $D + 1$)	17 parties	109 parties
Decontamination (D-day)	82 vehicles	12 vehicles

The initial ground-survey teams at the Nevada Test Site usually depart from the control point about 10 min after the detonation ($H + 10$ min). These teams have previously been briefed and have performed dry runs on their assigned access routes. Each shot area has eight radial roads, similar to the spokes of a wheel, starting from ground zero. The road is marked with stakes placed at 0.5 mile intervals so that the team location can be easily identified. As soon as the dust cloud settles, each team starts out from its IP (initial point) and obtains the location on the ground of the 10, 100, 1000 and 10,000 mr/hr line. The location of the dose rate line is referenced by odometer reading, to the nearest 0.1 mile, to the survey stakes. The readings are taken, using an AN/PDR-39 ion chamber instrument, from 10 to 15 ft from the vehicle to eliminate a possible shielding effect. The teams drive down one line obtaining the locations of the desired dose-rate lines, turn around and drive back a convenient distance, cross over to their next assigned line, and then survey this line in the same manner as their first line. As soon as these have been taken the survey teams withdraw to a location that is outside of the contaminated area. The results are radioed back to the control point as soon as possible. An aerial survey of pre-selected points is made by helicopter as soon as visibility permits. At the control point the readings are plotted on a large map of the area and dose-rate lines drawn for the 10, 100, 1000 and 10,000 mr/hr lines. These maps are used for briefing parties prior to their entrance into the contaminated area. Additional maps are prepared and placed at strategic locations throughout the test site. Resurveys of the areas are made on $D + 1$, $D + 2$, $D + 3$ and as frequently thereafter as required to maintain adequate surveillance of the areas. During "Operation Plumbbob", 432 area surveys were performed.

The health physics controls at the Nevada Test Site are as rigid as at Eniwetok Proving Grounds but are much more difficult to administer. Vehicle check stations are established

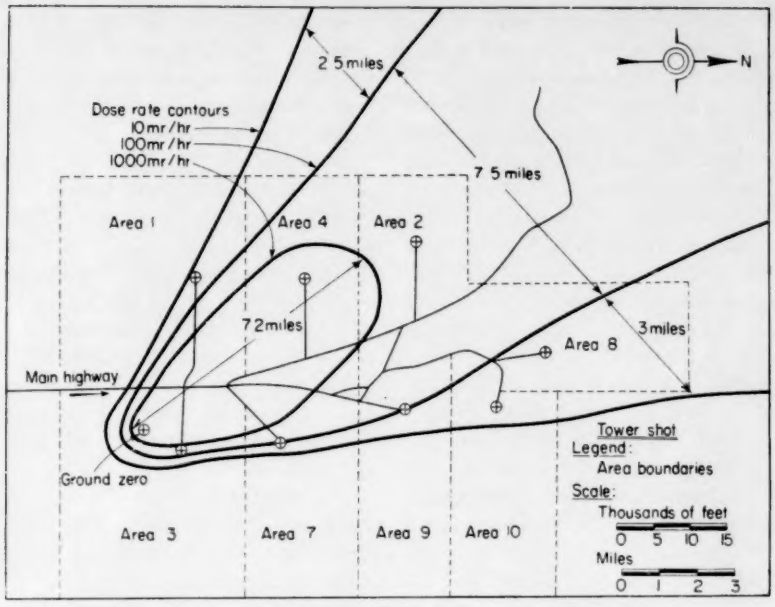

FIG. 1. Typical on-site fallout dose-rate contours for a tower shot.

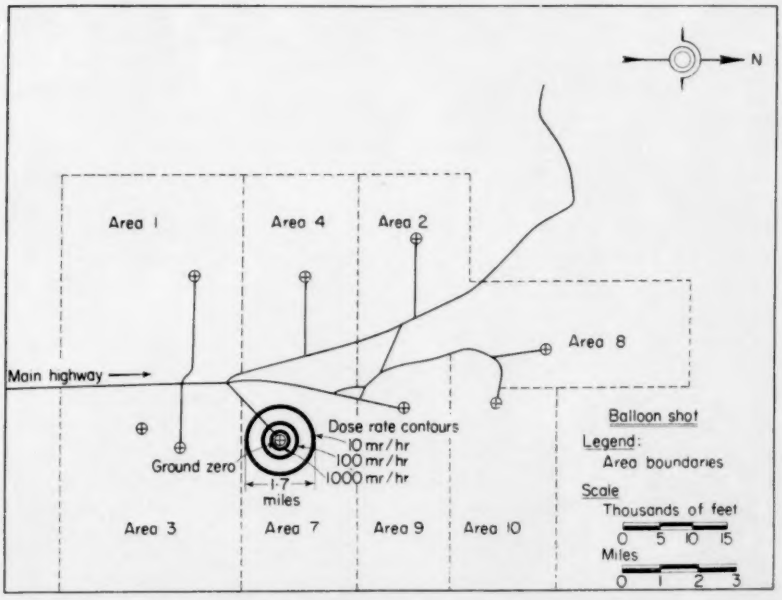

FIG. 2. Typical on-site fallout dose-rate contours for a balloon shot.

on the main access roads to contaminated areas. These stations cleared 25,600 individuals into the test area during the "Operation Plumbbob". At these stations all vehicles are stopped and the parties access permits checked to determine if all health physics regulations have been followed. A sticker reading "contaminated" and with the typical radiation warning symbol on it is placed on the windshield at this time. As the vehicle exits through the check station on its way out of the contaminated area it is monitored. If it is not found contaminated, the radiation sticker is removed. If contaminated, that is above 7 mr/hr, the party is directed to the vehicle decontamination station. Nearly 2000 vehicles required decontamination during "Operation Plumbbob". The check stations are moved frequently to maintain control of personnel entering contaminated areas but permitting routine activities to take place at other, non-contaminated, areas.

In a site as large as Nevada Test Site it is always possible for personnel to enter and exit from highly contaminated areas without passing a health physics check point. A roving monitor assigned to these areas can partially eliminate this problem. A lack of respect for radiation and inherent curiosity sometimes lead people into an undesirable situation. Several incidents have occurred at NTS in which the test participants have disregarded radiological safety regulations and warning signs. In particular, during a recovery operation in a high radiation field the recovery party would stay in the area longer than was permitted in order to complete collection of their technical data. Visitors to the test site have been known to enter radiation exclusion areas while engrossed in sightseeing activities. In either case, the individuals concerned could and have accrued radiation doses in excess of the maximum permissible exposure. Radiation warning signs and barricades are placed on the most frequently used roads to prevent personnel from entering unless properly cleared through the check point. The signs are moved periodically as the contamination decreases and/or increases due to a subsequent detonation.

Atmospheric, soil and water samples are collected from specific locations on a routine basis. These samples are analyzed for gross β–γ activity and α-activity. Swipe sampling, floor monitoring and various spot checks are performed in the living and working areas to ascertain that no contamination has been transferred from the foreward contaminated areas to clean areas. This program analyzed 10,000 samples during the operation; the negative results obtained indicated that no cross contamination occurred.

Various radioactive sources are required by project personnel for calibration, alignment, mock-up, or similar experiments. They are logged in when received, stored in a special storage area, and their history maintained while at the Nevada Test Site.

6. CONCLUSIONS

As in any health physics program, radiological safety in the weapons testing program becomes a routine procedure, but continues to require ever-present vigilance. The excitement and glamor usually associated with weapons testing is soon lost in the everyday task of providing health physics support and controls to the test organizations. Field operations are characterized by ever changing radiological situations and by the fact that radiation levels encountered are substantially higher than found in health physics work in laboratories. It is not at all unusual to furnish rad-safety controls for a balloon shot in the early morning, a tunnel shot at mid-morning, and a low tower shot in the afternoon. Consequently, the health physics controls become quite complex.

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IODINE MONITORING AT THE NATIONAL REACTOR TESTING STATION

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Abstract—Operations at the National Reactor Testing Station generally involve new types of fuel elements or reactors or the chemical processing of spent fuel elements. Because the isotopes of iodine are produced in high yield during fission and their short half-lives permit equilibrium concentrations to be built up rapidly, considerable inventories of these isotopes will be present after a reactor has operated for a relatively short length of time. The gaseous nature of the element allows its escape after rupture of the element cladding and particularly during chemical dissolution of short-cooled fuel. Iodine is one of the chief problems at this installation from the standpoint of radiological protection. The present paper describes some of the problems encountered and the way in which they were solved. Of particular interest is the development of a continuous monitor for gaseous iodine that permits immediate detection and records the buildup of activity as a function of time.

IODINE FROM RUPTURED FUEL ELEMENTS

INVESTIGATION of new types of experimental fuel elements may require that they be operated beyond the point of failure to determine maximum operating characteristics. The fission products released must be monitored positively and immediately for the protection of personnel and property in surrounding areas. Unless the test element was operated for a considerable time before failure, the isotopes of iodine will be of prime importance from the standpoint of radiological dose. There has been considerable uncertainty as to the physical state of iodine as released from a ruptured fuel element during operation. This question must be answered before adequate sampling measures can be devised for analytical control and may provide information that will be useful in the design of filters for air-cooled reactors.

During operation of a large reactor, a break occurred in the fuel assembly releasing fission products to the stack. Samples were taken directly from the stack, the air stream being drawn consecutively through two molecular membrane filters and two alkaline thiosulfate scrubbers, connected in series, at about 1 ft^3/min. The first filter was used to remove particulates from the gas stream, the second to indicate the efficiency of the first one. The first scrubber collects the gaseous iodine, and the second permits estimation of the scrubbing efficiency of the system. The millipore papers and the thick, porous cellulose papers used in back of the millipore for its support were dissolved in a chromic–sulfuric acid mixture to prevent loss of iodine. The resulting solutions and aliquots of the two scrubbers were then analyzed for iodine. Iodide was oxidized to the elemental state with nitrous acid which was then extracted into chloroform, stripped into alkaline sulfite and counted in a γ-well counter under well-defined and reproducible conditions.

Table 1 shows the distribution of iodine found in some typical experiments. The figures refer to the percentage of the total recovered iodine that was found in each fraction. In some experiments, the use of chemical fuels contributed a significant concentration of carbon in the stack as indicated roughly by the color of the first millipore filter. Gamma spectra indicated large quantities of gross mixed fission products on the first millipore filter in each case. With the exception of small quantities of radio-xenon in the scrubber solutions, only isotopes of iodine were present in the fractions beyond the first

Table 1. Percentage distribution of iodine from ruptured fuel element

No.	Color of first millipore	First filter		Second filter		Liquid scrubbers	
		Millipore	Support	Millipore	Support	No. 1	No. 2
1	white	5.7	2.5	0.9	0.9	87.7	2.2
2	light brown	7.3	2.0	1.1	1.3	86.1	2.2
3	black	27.0	4.7	1.4	1.5	63.9	1.5
4	dense black	60.7	15.3	0.5	1.7	21.3	0.5

millipore. The first scrubber contained most of the iodine when the carbon loading was not excessive and the efficiency of the liquid scrubbers was 98 per cent in each case. The iodine activity of the second millipore and both support papers is attributed primarily to adsorption of the gaseous molecule except for the first support paper in test (4). This paper was slightly yellow and obviously had trapped some very finely divided carbon that had penetrated the first millipore paper.

Apparently, iodine released from an operating fuel element at high temperature is predominantly gaseous, and its collection on a mechanical filter will depend on its having been adsorbed on a dust particle during its previous history. Iodine gas is partly adsorbed by anything with which it comes in contact and will be found on metals, glass, rubber, plastics, paper, etc. However, the process is very inefficient as a quantitative method of collection. Many of the conventional methods of monitoring for airborne radioactive materials such as collection on filter paper or fallout plates are obviously inadequate because of the small surface area presented to the gas stream. The data of Table 1 shows only 1–2 per cent of the gaseous iodine was collected on either a millipore or the thick cellulose support paper.

IODINE LIBERATED DURING DISSOLUTION OF SPENT FUEL ELEMENTS

When a spent fuel element is dissolved in nitric acid at the beginning of chemical processing for the recovery of the unfissioned fuel, those fission products that are gaseous under the conditions employed will be evolved in the off-gas stream. Chief among these will be iodine and the inert gases, xenon and krypton. In the Rala process for recovery of barium-140, spent fuel elements are dissolved after having been cooled for only a short period. Consequently, large quantities of essentially pure isotopes of iodine will be released from the nitric acid solution. Unless the iodine is very thoroughly scrubbed from the gas stream before release to the stack, the iodine will become a distinct radiological hazard to people working at short diffusion distances from the stack both inside and outside the plant.

In order to monitor iodine released during Rala operations, liquid scrubbers were again used because of their good efficiency for collection of iodine gas and because the solutions are more convenient to analyze than with other methods of collection. However, the scrubber used in the earlier work was improved by using a 6-in. disc of fine-porosity stainless steel as the air diffuser. The efficiency of the new scrubber was checked in the laboratory by passing iodine gas generated from iodine-131 through the scrubber at a flow rate of 1.5 ft^3/min, and was found to be better than 99.9 per cent. Samples were then taken directly from the Chemical Processing Plant stack during a Rala run using two millipore filters and two scrubbers in series as before. To elimate any complications from rare gases or their daughter products, iodine was separated chemically under conditions known to give complete recovery of ionic types of iodine. The procedure includes extraction into chloroform, stripping back into aqueous solution and precipitating as palladous iodide. The precipitates were β-counted under reproducible counting conditions. Contrary to expectations, the scrubbing efficiency was very poor. The

precipitates contained 56,870 c/m and 39,750 c/m from the first and second scrubbers, respectively. The scrubbing efficiency is thus only 30 per cent as compared to 98 per cent obtained with iodine from a ruptured fuel element with a less efficient scrubber, and to 99.9 per cent for the same scrubber with known molecular iodine. Part of the iodine evolved in the Rala process evidently is not present as iodine molecule but as some less chemically reactive form that is only partly retained by alkaline thiosulfate. Furthermore, the precipitates decayed at markedly different rates, the first decreasing 59 per cent and the second 83 per cent during the same 25-hr period. Resolution of the decay curves showed unmistakably the presence of only 8-day iodine-131 and 2.3-hr iodine-132 (from 77-hr tellurium-132) in both precipitates. Iodine-133 and iodine-135 were rarely observed because of decay during cooling. The presence of both iodine-131 and iodine-132 in the second scrubber indicates that both isotopes are involved in the new chemical form. Moreover, the higher ratio of iodine-132 to iodine-131 in the second scrubber indicates greater production of the new form and even poorer scrubbing efficiency with the heavier isotope. The same experiments have been repeated many times, and while there have been quantitative differences due to differences in time of analysis or sampling, the results always show very poor scrubbing efficiency and faster decay in the second scrubber. Other evidence indicates that more than one new form may be present.

One of the most significant differences between the release of fission products from a ruptured fuel element and from an element after dissolution in nitric acid is the copious evolution of nitric oxide and nitrogen dioxide that accompanies the latter. Both molecules have single unshared electrons. Since the isotopes of iodine are produced atom by atom each of which has an unshared electron, compounds of the type NOI and NO_2I are possible. Because of the greater rate of formation and decay of the I^{132} than I^{131} during the dissolution process, it is likely that more of the former atoms would be converted to the covalent type. The effect of such molecular compounds of iodine in producing internal dose to the body is not known but it is expected to be small because of its low chemical reactivity.

FIELD EVALUATION OF GASEOUS IODINE

Another iodine monitoring problem that is generally present at the NRTS has an important economic aspect in addition to the normal one of personnel protection. Frequently, large numbers of people may be employed on maintenance or construction work in the vicinity of an operating reactor. During experimental operations, possibility exists for some of these people to receive a significant dose to the thyroid from inhalation of iodine gas. The problem is particularly acute for workers in the vicinity of the Chemical Processing Plant stack during a Rala operation. The evacuation of one such group for a few hours once a month just for the period of operation was estimated to cost nearly one hundred thousand dollars a year. On the other hand, to permit them to work during the operation and determine after-the-fact the thyroid dose received might result in serious injury or expensive lawsuits. The use of liquid scrubbers is obviously not a suitable answer. The time required to take an adequate sample, return it to the laboratory and complete an analysis, and get instructions back to the field is too long for this technique to be of much help to people accumulating iodine all the while.

Continuous monitor

A continuous monitor has been devised for use where relatively large concentrations of people are present in sensitive locations. The device gives an immediate indication of the presence of iodine activity, keeps track of the total accumulation of iodine with time, which is proportional to the build-up of iodine in the thyroid gland, and presents the data as a permanent written record for either legal or scientific purposes.

The heart of the device is shown in Fig. 1. The monitor cartridge is constructed from a flexible acetate plastic vial, $\frac{5}{8}$ in. diameter $\times$ $2\frac{1}{2}$ in long. A 4-in. length of $\frac{3}{16}$-in. o.d. brass tubing with the lower end cut at a 45° angle is slipped through a $\frac{3}{16}$-in. off-center hole in a $\frac{5}{8}$-in. circle of 60-mesh brass screen. A piece of $\frac{5}{8}$-in. Tygon

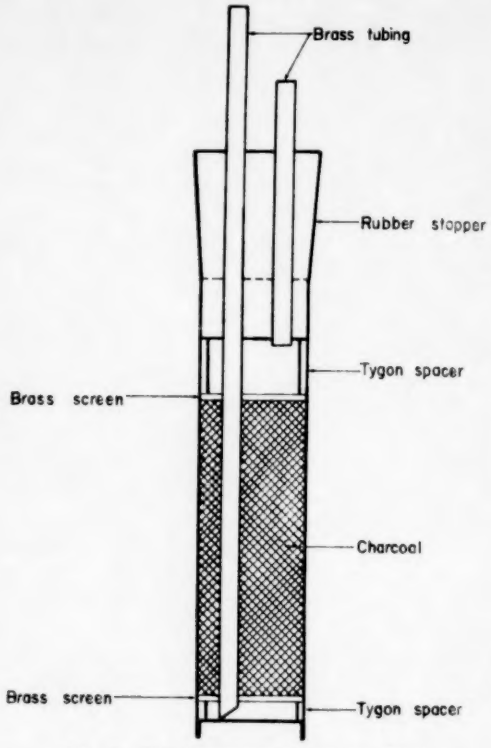

FIG. 1. Continuous-monitor cartridge.

tubing about $\frac{3}{16}$ in. long is placed in the bottom of the plastic vial. The tube and screen are lowered into the vial until the screen rests firmly on the spacer and the tube rests on the bottom of the vial. The vial is filled with 12 × 30 mesh activated carbon. Another screen with the hole a little off center is slipped down the tube and pressed tightly against the top of the carbon column and another Tygon spacer added to hold the screen in place. A two-hole No. O rubber stopper is slipped down over the tube until it fits tightly in the vial. Another $1\frac{1}{2}$-in. piece of $\frac{3}{16}$-in. o.d. brass tubing to which the pump is to be attached is inserted in the other hole of the stopper.

In actual monitoring practice the cartridge is placed in a lead-shielded, thallium-activated sodium iodide well-counter connected to either a linear or logarithmic counting-rate meter. The output of the counting-rate meter is fed to a recording instrument which plots activity as a function of time. Thus a permanent record is obtained of the activity build-up in the cartridge as a function of time. A schematic diagram of the continuous monitor is shown in Fig. 2.

Field cartridge

A field cartridge is designed for monitoring outlying field locations where time is not so important and where the large number of sampling stations required will not permit the expensive auxiliary equipment used with the continuous monitor to be duplicated at every station. A $\frac{1}{4}$ h.p. pump to draw air through the cartridge is the only auxiliary equipment required, the cartridge being simply inserted in the end of the tubing leading to the pump. The cartridges can be picked up and returned to the laboratory for analysis or they can be counted in the field by making the rounds of the sampling stations with a mobile counter.

The cartridge shown in Fig. 3, is composed of a cylinder of flexible acetate plastic, $\frac{5}{8}$ in. in diameter by 2 in. in length open on both ends. The cylinder is filled with 12 × 30 mesh activated carbon with $\frac{5}{8}$-in. circle of 60-mesh brass screen at each end to hold the carbon in place, and a piece of $\frac{5}{8}$-in. Tygon tubing about

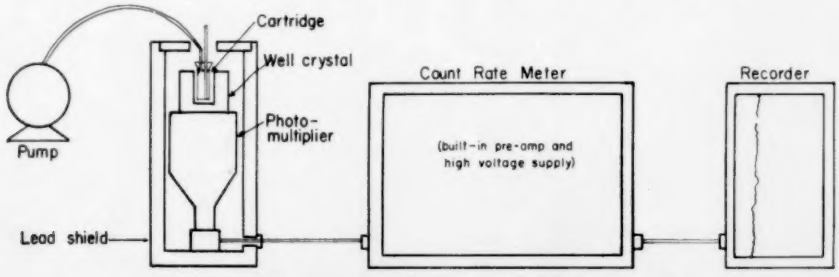

FIG. 2. Schematic diagram of continuous monitor.

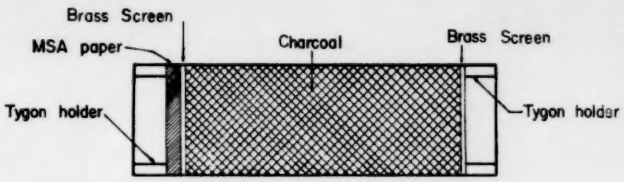

FIG. 3. Field cartridge.

$\frac{3}{16}$ in. long as a retainer ring to hold the screens in place. A $\frac{5}{8}$-in. circle of MSA 2133 all-dust paper is inserted at one end between the screen and retaining ring. The filter can be removed and the particulate activity counted separately from the iodine if desired. The screens and filter are stamped out easily in quantity with a $\frac{5}{8}$-in. punch and the retaining rings can be snipped off a length of Tygon tubing with a pair of scissors. The total cost of the cartridge including both materials and labor is about 8 cents. These cartridges have replaced devices of the liquid-scrubber type employing a 6-in. diameter, porous stainless steel diffuser in a stainless steel can costing about $75 each. Even greater savings result in terms of manpower-hours required to make analyses starting from the compact carbon cartridges compared to large volumes of solutions. The cartridge is designed to be placed directly in a standard $\frac{5}{8}$-in. thallium-activated sodium iodide well counter and counted with or without the filter containing the particulate activity. Larger models can be made when greater sensitivity is desired. Two other real advantages resulting from use of the cartridge are elimination of the freezing of solutions during cold weather and evaporation of the solvent during hot weather that occur with liquid scrubbers.

Table 2. Adsorption of iodine on activated carbon

A. Laboratory Run

$\frac{1}{4}$-in. section	Activity found (c/m)
1	58,900
2	4600
3	490
4	80 ± 27
5	0 ± 25

ADSORPTION OF IODINE ON ACTIVATED CARBON

The collection efficiency of activated carbon for gaseous radioiodine was determined in the laboratory for molecular iodine and on samples taken from the Rala stack for whatever species might be present. The data are listed in Table 2. The sampling rate in all tests was from 1 to 1.5 ft^3/min. In part *A*, iodine was generated from Oak Ridge iodine-131 and drawn through a single field cartridge divided into five sections each approximately $\frac{1}{4}$ in. in length. The efficiency is excellent considering the small quantity of carbon and relatively high flow rate. In part *B*, the off-gas from the plant was sampled through two of the field cartridges in series during dissolution of a fuel element. The gas stream had been prefiltered through two millipore filters. Although some break-through might have occurred, both efficiency and capacity are obviously adequate for sampling purposes. In part *C*, a piece of 25-mm Pyrex tubing was filled with activated carbon using 60-mesh screens to divide the column into six sections each $5\frac{1}{8}$ in. long and containing 12.5 g of carbon. The air stream was drawn through a series train consisting of two millipore filters and their support papers, the carbon column and finally two alkaline thiosulfate scrubbers using the 6-in.-steel air diffuser. Decay curves were then determined for each section by counting

B. Plant Stack

Run no.	Activity found (c/m)	
	Cartridge 1	Cartridge 2
1	18.9×10^6	18.7×10^4
2	24.1×10^6	15.4×10^4
3	1.2×10^6	1.2×10^4

C. Plant Stack

Section	Activity found (c/m)	Remarks
Millipore and Support paper		
1	164,082	>90% I^{131}
2	13,933	
Activated carbon		
1	12.5×10^6	95% I^{132}, 5% I^{131}
2	1.9×10^6	83% I^{132}, 17% I^{131}
3	1081	
4	1017	100% I^{131}, no I^{132}
5	976	
6	928	
Liquid scrubbers		
1	27,900	100% I^{131}, no I^{132}
2	11,200	

D. Field Samples

$\frac{1}{4}$-in. section	Activity found (c/m)	
	Sample 1	Sample 2
1	198 ± 11	6480 ± 37
2	3 ± 10	892 ± 17
3	16 ± 10	328 ± 12
4	14 ± 10	298 ± 12
5	15 ± 10	132 ± 11
	246 ± 23	8130 ± 45

in a γ-well counter. Most of the activity on the first two carbon sections decayed with a half-life of 2.4 hr followed by a clear 8-day tail. The last four carbon sections and the two scrubbers showed a clean 8-day decay with no evidence of shorter-lived activities.

Because of small variations in the counting efficiency, the percentage figures are not quite exact but the qualitative conclusions seem reasonably clear. There is probably more than one chemical form of iodine present in the Rala off-gas that is significantly different in its action than iodine molecule. The poor collection efficiency through the last four carbon sections and the two liquid scrubbers indicates a different form for iodine-131. The distribution of iodine-132 in the first two carbons and its complete absence in succeeding fractions may indicate yet another form whose collection on carbon is more efficient than with the iodine-131 form. This sample was obtained 2 days after the Rala operation was completed. It is possible that the odd iodine-132 molecules might have decayed and have been replaced by normal iodine-132

from decay of tellurium-132. However, another sample taken a day later showed the same abnormal distribution of I^{132} in liquid scrubbers. The collection efficiency for either form is apparently better with activated carbon than with alkaline thiosulfate scrubbers since even with iodine-131, a considerable quantity of which was present in the scrubbers, more than 95 per cent of the total was present in the carbon. Because of inability to control conditions in the stack, each sample may differ considerably from another in the way it acts. The data presented should not be considered as final but as an indication of the types of samples that have been encountered. Much additional work needs to be done to determine the mechanism involved.

CALIBRATION

Calibration of either carbon cartridge is relatively simple. A 1-ml aliquot of standard iodine-131 solution is placed in one of the $\frac{5}{8}$-in. plastic vials and counted in the well counter to be used. A spacer can be used if desired to raise the vial of standard iodine slightly so that the bottom of the solution will be at the same height above the bottom of the well as the bottom of the carbon bed. However, since the well crystal is $1\frac{1}{2}$ in. deep but only $\frac{5}{8}$ in. in diameter, the counting efficiency for iodine-131 changes relatively by less than 3 per cent as the depth of solution changes from zero to about $\frac{1}{2}$ in. Most of the iodine activity in an air sample will be collected in the first $\frac{1}{4}$ in. of the cartridge, even at levels greater than will normally be encountered in field work (Table 2, *A*). Because the counting efficiency also depends on both the γ-energy and the decay scheme of the particular isotope, the above calibration will not be correct for other isotopes or mixtures except in fortuitous circumstances. Accordingly, the MSA prefilter must be removed from the field cartridge before counting, unless activities other than iodine-131 are absent. Similarly, quantitative values obtained from the continuous monitor may be in error considerably if other activities are present. The air stream can be prefiltered, or the monitor can be used as an indicator and quantitative answers obtained by γ-spectroscopy on the carbon cartridge.

SENSITIVITY

The sensitivity of detecting radioactive iodine present in the cartridge will be dependent mainly on counting time and instrument background. If air concentrations are desired, sampling rate and time are also necessary. Consequently, sensitivity will vary depending on the actual conditions used. Thus, for a 5-min count on both sample and background with a background of 230 c/min the detection limit is 20 ± 10 c/min and is defined as the net count that is equal to two standard deviations of its own determination. With a counting efficiency of 40 per cent for iodine-131, the detection limit amounts to about 50 d/min or 2.2×10^{-5} μc. For a sample taken at a rate of 1 ft^3/min for 1 hr, the detection limit in terms of air concentration would be 1.3×10^{-11} μc/ml. A sensitivity figure can also be given in terms of thyroid dose, if it is assumed that the standard man at work breathes at a rate of 0.7 ft^3/min and 15 per cent of the inhaled iodine is fixed in the thyroid gland. The detection limit of 50 d/min for a 5 min count at a sampling rate of 1 ft^3/min would correspond to 1.4×10^{-5} rad for an individual breathing air of the same iodine concentration for the same period of time. Longer counting times or larger samples will obviously result in greater sensitivity.

The calculations above assume that all the iodine in the air stream is collected by the carbon cartridge. That this is true for concentrations giving 64,000 c/min during a collection period of about 30 min is shown by the laboratory data of Table 2, *A*, and for even higher concentrations and short sampling periods in parts *B* and *C*. However, it is particularly desirable to know if the cartridge will collect iodine efficiently at concentrations near the detection limit, i.e. at concentrations so low that little activity is collected during many hours of sampling. The data of Table 2, *D* show the excellent distribution of iodine through two field cartridges that were used to monitor a Rala operation. The activity was shown by γ-spectra to consist entirely of iodine-131 most of which is concentrated in the first $\frac{1}{4}$ in. Both samples were collected for 96 hr at a rate of 1 ft^3/min at diffusion distances of 300 and 600 ft from the top

of the stack. Although the iodine concentration in the air undoubtedly did not remain at its average value throughout the entire sampling period, the concentration must have been extremely small at any time. Iodine was known to be leaving the stack throughout most of the period, and dilution would have been considerable. If the iodine had been adsorbed only during momentary passage of a cloud of relatively high concentration, subsequent movement of air through the cartridge for up to 96 hr would desorb the iodine completely or move it further down the cartridge unless the efficiency was high, even for low concentrations.

In regard to calibration of the cartridge, it is interesting to note that sample No. 2 had a counting rate of 7885 ± 40 before being disassembled for the distribution study. The counting rate is thus increased by only 3 per cent when each section is counted in the bottom of the well rather than being stacked on top of each other as in the normal distribution.

LARGE WELL CRYSTAL FOR γ-COUNTING

Another monitoring tool that has proved to be very useful is a large thallium-activated sodium iodide well crystal. This is similar to the standard $\frac{5}{8}$-in. well except that it is made from a 3 in. $\times$ 3 in. crystal with a well 2 in. in diameter and 2.5 in. deep. About 100 ml of sample can be counted with an efficiency of about 40 per cent in most cases and a background of about 400 c/min. The large wells have been particularly useful in screening samples of urine, milk, vegetation, soil, etc. for radioiodine. The volatility of this element makes any chemical treatment or concentration rather complicated, and the time required allows decay of shorter-lived isotopes. Since relatively large samples can be used without preparation of any kind a very fast method of screening samples is available. Of course, if activity is found recourse must be made to chemical separations or γ-spectroscopy for identification and determination. Many other uses are obvious. Various organs of interest from experimental animals can be screened quickly for γ-activity. During one field release test in which rats were used, they were screened by putting their top half into the well. Absence of any radioactivity made it unnecessary even to dissect the organs of interest in a great number of cases.

THYROID DOSE FROM URINARY EXCRETION

Several individuals were exposed to radioiodine gas accidentally during a Rala operation. The dosage was not high enough to cause any detectable biological damage but did provide some excellent samples. Samples were taken from each individual over a period of about 30 days and analyzed for iodine-131. The excretion curves in each case showed two well-defined straight lines on a semi-log plot. Excretion during the first few days was very rapid with an effective half-life of about 7–8 hr. After 4 or 5 days the slope of the curve changed to one with an effective half-life of about 7.5 days and continued at this slope until sampling was terminated. It was felt that the rapid elimination observed during the early part of the curve represented iodine that was not fixed in the sense that it was organically-bound in the thyroid gland. Direct measurement of the thyroid gland made by Phillips Petroleum Company with a thyroid counter on the same men at the same time showed a single straight line with effective half-life of about 7 days. Calculation of the thyroid dose gave results that were in surprisingly good agreement with those obtained from direct measurement of the thyroid if only the latter part of the curve was used.

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IN VIVO GAMMA COUNTING AS A MEASUREMENT OF URANIUM IN THE HUMAN LUNG

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Abstract—A direct, *in vivo* method is reported for the measurement of uranium within the human lung, utilizing the detection by scintillation spectroscopy of the gamma radiation accompanying alpha decay of the uranium. The *in vivo* gamma counting facility includes a low background counting chamber with 8-in. thick iron walls, NaI scintillation detector, and a multichannel pulse-height analyzer. To achieve a quantitative estimate of uranium concentration, two techniques for estimating the normal, non-uranium gamma activity at the characteristic uranium gamma energies have been developed. Calibrations have been derived from measurements of samples in phantoms simulating the human chest. The 186 keV gamma line from U^{235} is the most suitable for measurement, but if secular equilibrium is assumed, the 90 keV lines of the uranium daughters may be used. The ratio of the 90 keV line to the 186 keV line may be used as an indication of U^{235} abundance. Quantities as low as 50 per cent of the maximum permissible body burden of any of the isotopic enrichments investigated can be measured.

INTRODUCTION

TYPICAL safety precautions for the industrial processing of radioactive or otherwise hazardous materials consist of the procedures of environmental monitoring of the process areas, to insure that the area and air contaminations are held at prescribed safe levels, and examinations or monitoring of personnel involved in the processes. The mode of personnel monitoring previously used for uranium processing has been excretion analysis, usually urinalysis, for uranium elimination from the body. The desirability of a direct method of measuring the concentration of any isotope in the human body as a supplement to and a cross-check on the less direct excretion analyses, is obvious. These benefits are particularly appealing for insoluble radioactive materials, such as uranium and some of its compounds, which may be inhaled and are excreted only by complex, indirect modes.

The recently developed techniques of scintillation spectroscopy of the gamma radiation from the human body have been used to measure the gamma radiation of uranium and thence the uranium content of the human lung, as described in this paper.

In the course of planning this method development program and constructing the required facilities, a request was made of the *in vivo* gamma counting group at Argonne National Laboratory to attempt such a measurement on a Y-12 employee involved in a uranium inhalation accident. The successful lung burden measurement made on this employee (Case Y-1) through the courtesy of MILLER, MARINELLI and ROSE[1,2] of that laboratory was a validation of the Y-12 feasibility studies for the program and a useful guide for certain aspects of the development of the routine method.

THE *IN VIVO* GAMMA COUNTER

General

The Y-12 *In Vivo* Radioactivity Measurement (IVRM) Facility, in which the measurements have been made, is basically a gamma counter consisting of a large-volume scintillation crystal,

* Operated by Union Carbide Nuclear Company for the United States Atomic Energy Commission.

equipment for gamma energy distribution studies by pulse-height analysis, and a low-background counting chamber large enough to contain conveniently the person being examined and the scintillation detector.

The Y-12 IVRM Facility is essentially of the Argonne National Laboratory type which has been adequately described previously.[3] Therefore, only a brief description of the facilities, with emphasis on unusual features and problems, is included here.

Scintillation detector

The gamma scintillator used in the *In Vivo* Radioactivity Measurement Facility is a 9-in. diameter by 4-in. thick thallium-activated sodium iodide crystal. The crystal is packaged in electrolytic copper with a quartz window. The electrolytic copper was recommended by the manufacturer and others as having lower radioactivity than the aluminum normally used and the quartz window as reducing the background activity (particularly K^{40}) usually contributed by a Pyrex window.

As indicated in Fig. 1, a 5-in. diameter photomultiplier tube is coupled directly to the crystal window, using silicone fluid (viscosity = 10^6 cS) as the optical couplant. The area of the crystal window not covered by the photomultiplier is covered with a machined Teflon reflector. A single tube cathode follower-type preamplifier transmits the pulses from the photomultplier to the counting equipment, which is located outside the counting chamber.

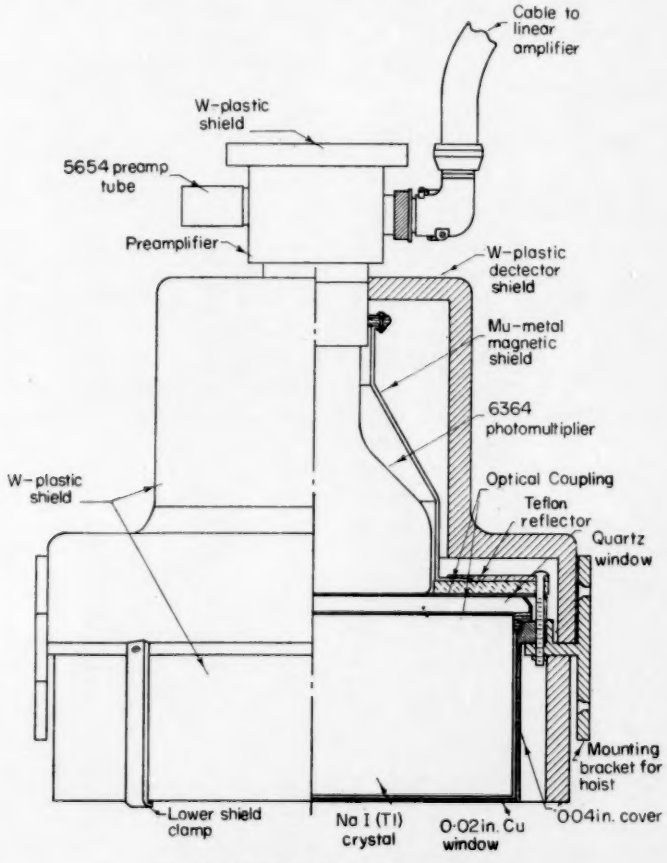

FIG. 1. IVRM scintillation detector.

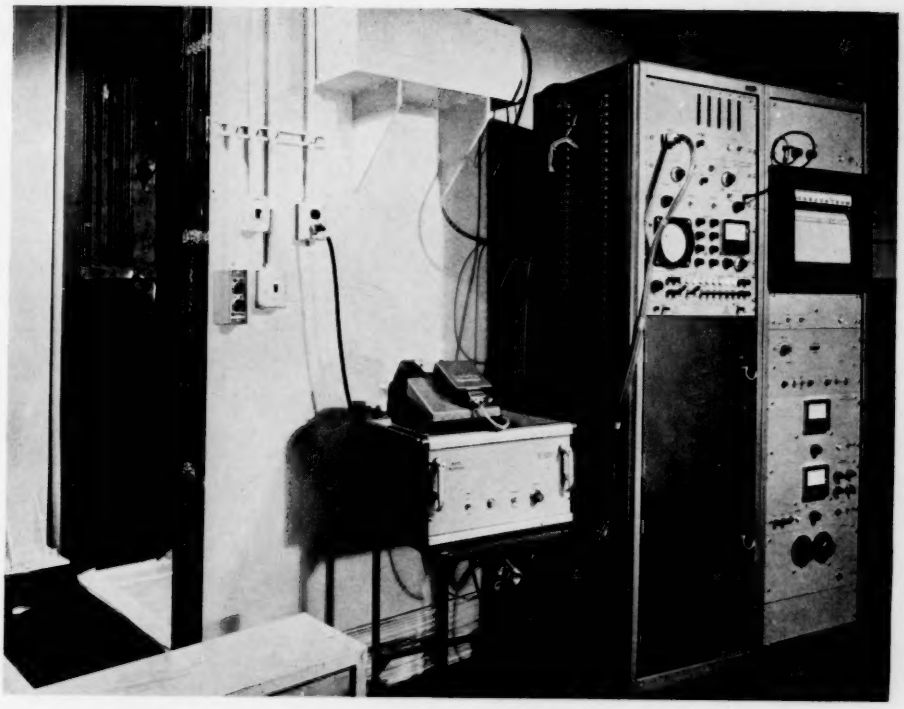

FIG. 2. Multichannel pulse-height analyzer.

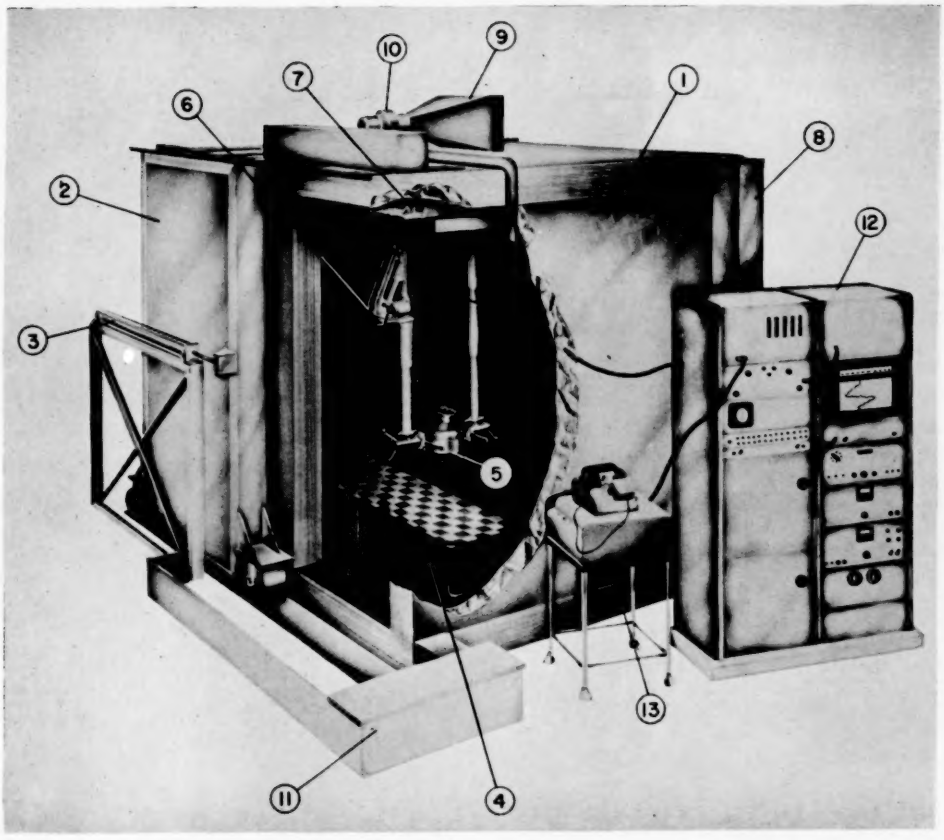

FIG. 3. Y-12 low-background gamma counting facility. (1) Laminated steel walls, roof and floor—32 sheets ¼-in. thick iron. (2) Door (3) Pneumatic cylinder—door drive (4) ¼-in. thick tungsten–plastic shielding (5) Shielded scintillation detector (6) Detector hoist (7) Detector bridge and carriage (8) Steel grit enclosures (9) Air-supply filter (10) Air-supply blower (11) Entry area fence and seat (12) 256-channel pulse-height analyzer (13) Digital recorder.

The detector is shielded by a ½-in. thick cover of tungsten-powder-loaded epoxy plastic in all areas except the front face. Among other things, this shield helps limit the radiation acceptance angle of the detector, reducing the effect on the detector of radiation from body areas other than that under examination. This shielding material has a density of 9–11 g/cm^3 and a linear absorption coefficient equal to about ½ that of lead for 660 keV radiation. The material is used because of its very low radioactivity and good shielding characteristics.

The detector is suspended from a hoist arrangement that permits it to be raised or lowered over a range of 24–50 in. from the floor of the counting chamber. A bridge and carriage permits movement of the detector in a horizontal plane.

Multichannel pulse-height analyzer

A 256-channel pulse-height analyzer is used to amplitude-sort the pulses from gamma scintillations in the detector and provide the energy distribution spectrum. The magnetic core memory of the instrument has a count storage capacity of 65,535 counts per channel. Although the analyzer has provisions to correct for dead-time losses, the normal gamma activity of the human body is too low to require this correction.

The information stored in the instrument may be displayed on a cathode ray tube or a strip chart recorder as an actual counts-as-a-function-of-energy distribution or printed on paper tape as a digital presentation of the number of counts in each of the 256 channels. The latter mode is the quantitative read-out, using an acumulation-type adding machine as a digital printer. The printer produces a sequential recording on the tape of an identification number and the number of counts for each channel and also the sum of the number of counts in adjacent 8 channel bands. For high resolution of gamma ray energies, the information in the 256 individual channels is useful. For routine gamma spectrometry of known activities in which the area under sections of the spectrum is used for quantitative measurements, the 32 band spectra made up of the sums of each 8 adjacent channels minimizes integration and reduces data handling.

The 256-channel analyzer is shown in Fig. 2.

Low-background counting chamber

The low-background counting chamber built was modeled after the "iron room" shielding developed at Argonne National Laboratory.[3] In the Y-12 Iron Room, 8 ft × 8 ft panels of ¼-in. thick steel for the laminated walls, floor, ceiling and door were used. The Iron Room measures 6 ft 8 in. × 6 ft 8 in. × 7 ft 6 in. high inside, with 32 sheets of ¼-in. thick hot-rolled steel sheets in each of the 8 in. thick walls, the floor, door and ceiling. The construction is "platform" type, with the vertical walls built around an angle iron frame on the floor. The ceiling overlaps the vertical sides to block direct radiation entry paths into the chamber. The sheets were clamped in place and tack-welded. Efforts were made to keep welding inside the room to a minimum. The cracks at each corner, where the side wall form 90° angles, are covered with 8 in. × 8 in. sheet-metal enclosures filled with steel grit. The door of the room provides a lap closure around the door opening. This door moves on 7½-in. diameter × 6-in. wide steel wheels, and is driven by a pneumatic piston. Push-button switches inside and outside the room operate the door.

A low wooden fence and bench provide an isolation or transition area at the door. This device minimizes the possibility of contamination of the interior of the low-background chamber.

Conditioned air is drawn from the room in which the counting chamber is located, through a CWS-6 filter, and distributed into the Iron Room.

The Y-12 Iron Room is shown in Fig. 3.

In addition to the gross iron shielding, thin lead linings have been used within iron rooms to reduce scattered radiation caused by radiation from the residual activity in the detector or in the subject striking the iron walls.[4] Initially, the Y-12 Iron Room was lined with ⅛-in. lead. Comparative measurements of the background of the bare and lined room and of samples of the lead suggested that some activity was included in the lead. After considerable

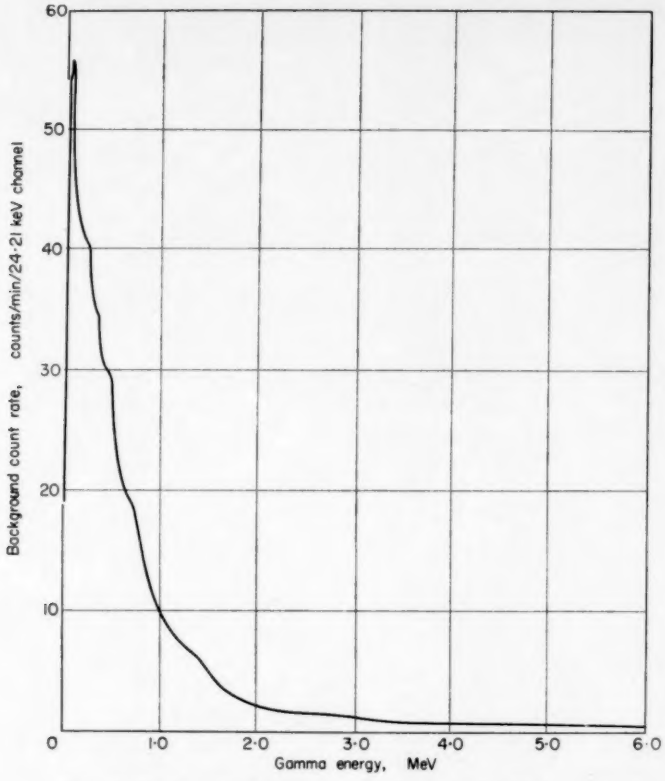

FIG. 4. Background gamma spectrum in Y-12 iron room

testing with shielding adjacent to the detector, it was decided that the background could best be reduced by lining the room with tiles of the tungsten-loaded plastic mentioned above. The floor and the lower portion of the walls (to a height of 35 in.) were covered with the tiles and the uncovered portions painted with clear Tygon paint to prevent rusting. The detector shielding described in the *Scintillation detector* section was designed at this time to reduce the radiation from above that might strike the detector.

Fig. 4 shows the present background of the Y-12 Iron Room to 6 MeV.

Operation

For the work discussed in this paper, the objective of most of the *in vivo* radioactivity measurements was detection of deposits of uranium in the human chest. For this purpose, the measurement is usually made with the subject supine on a cot, with the open face of the scintillation detector in contact with the chest. In special circumstances, measurements have been made in the whole-body positions (1-m arc and tilted chair) described by EVANS, MARINELLI and MILLER[5–7].

Shower and locker facilities are located near the *In Vivo* Radioactivity Measurement Laboratory to permit the subjects to shower thoroughly and change into radioactively clean clothing for the measurement.

To facilitate any possible correlation between uranium urinary excretion and IVRM data, a urine sample is frequently requested of the subjects at the time of the examination.

Information regarding name, badge or identification number, work area, job description, weight, height, age, sex, chest thickness, chest width and abdominal thickness is taken for

each subject and recorded on one side of a file card which is prepared for every counting run. The reverse side of this card serves as a convenient place to record the 32 bands of spectral data from the pulse height-analyzer.

An operator is always stationed outside the Iron Room during examinations of persons. This procedure permits the operator to process data taken from previous examinations, to periodically observe the accumulating spectrum, and to monitor the subject's communication channel through the microphone in the Iron Room.

In routine operation, the facility time required for each *in vivo* gamma examination has been found to be the desired counting time plus about 10 min for entry and exit from the Iron Room, positioning of subject and detector, and digital recording of the gamma spectrum from the multichannel analyzer.

PRINCIPLES AND TECHNIQUES OF METHOD

Gamma activity from the MPBB amounts of uranium

The actual radiation hazard from inhalation or ingestion of uranium is due to the energy dissipated in small volumes of tissue by the alpha particles emitted. Direct, *in vivo* measurement of uranium depends, however, upon the gamma emission accompanying the alpha decay, since the alpha particles do not penetrate the body wall. For this reason, it is necessary to consider the gamma activity associated with the maximum permissible body burden (MPBB) amount of uranium.

The International Commission on Radiological Protection recommends 0.01 μc as the MPBB for insoluble natural uranium, citing the lungs as the critical organ.[8] By definition, the curie of natural uranium in the recommendation has special meaning as follows:

$$\text{special curie} = \begin{cases} 3.7 \times 10^{10} \text{ dps (disintegrations per second) } U^{238} \\ 3.7 \times 10^{10} \text{ dps } U^{234} \\ 9.0 \times 10^{8} \text{ dps } U^{235} \end{cases}$$

Since the special curie amounts to 7.49×10^{10} alpha dps/c, the MPBB of natural uranium is 749 alpha dps.

Current health physics problems include not only natural uranium, but also mixtures of uranium isotopes in which the U^{235} content varies to greater than 90 per cent. Moreover, the processes of U^{235} enrichment concentrate the U^{234} in the natural material along with the U^{235}. Because of its high specific activity, the U^{234} becomes the major contributor of alpha activity as its concentration in uranium increases. Also, as the relative contributions of the various uranium isotopes to the total alpha activity change, the average energy of the total activity varies because of differences in the energies of alpha emission from the isotopes (4.2 MeV for U^{238} alphas, 4.7 MeV for U^{234}). To summarize, each uranium isotopic concentration level will have its own MPBB in terms of weight of uranium and is subject even to slight change in the number of alpha dps/MPBB.

In Table 1 are listed three isotopic enrichment levels of uranium, the typical weight concentration of U^{235}, and the MPBB weight calculated for each level from the isotopic analysis and the ICRP recommendation, as described above.

The *in vivo* gamma counting method is handicapped because of the heavy proportion of alpha particles from U^{234}, which does not contribute to the gamma spectrum. The only prominent features of the uranium gamma spectrum are lines at 186 keV and about 90 keV. The 186 keV line is from U^{235}, with an abundance of 0.8 γ/α. The approximately 90 keV lines are due to Th^{231}, the daughter of U^{235}, and to Th^{234}, the daughter of U^{238}. These daughters have different but rather short half-lives (25 hr and 24 days, respectively) and so will be in evidence after chemical purification as they approach secular equilibrium with the uranium parents. The gamma quanta from U^{238} and U^{234} themselves essentially all undergo electron conversion and the resultant 10–20 keV X-rays are too heavily absorbed by the body wall to be of use in the *in vivo* counting method. It may be concluded that the *in vivo* counting method should utilize the 186 keV line to determine U^{235} content when its abundance is sufficiently high. The relative magnitude of the 90 keV line is an approximate indicator of the isotopic concentration levels. Use of the 90 keV lines

Table 1. *MPBB amounts and gamma activity of three uranium isotopic enrichment levels*

Level	Description	Typical U^{235} content (Weight %)	MPBB Uranium (mg)	MPBB U^{235} (mg)	Gamma activity 186 keV (γ/s/MPBB)
1	U^{235}-enriched	93.0	0.275	0.255	16.1
2	Natural U^{235} abundance	0.7	25.615	0.180	11.4
3	U^{235}-depleted	0.22	49.624	0.109	6.9

for evaluation of uranium body burden would be subject to errors because of uncertainties regarding the equilibrium conditions between parent and daughter. Therefore, Table 1 also lists the 186 keV gamma activities to be expected from the MPBB of each level of uranium-235 enrichment.

Fig. 5 is the relative spectra in air of the MPBB amounts of the three levels as taken with the IVRM detector.

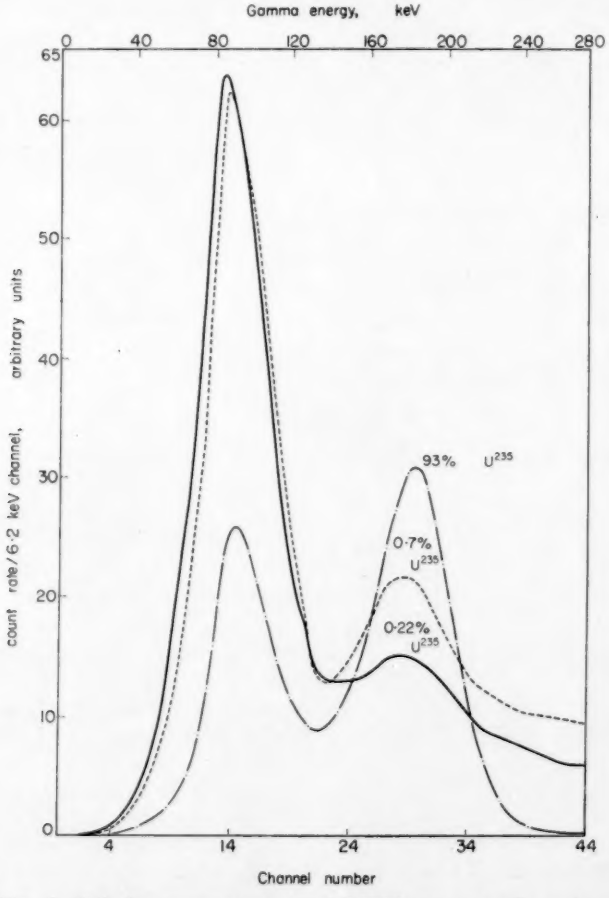

FIG. 5. Relative gamma spectra of MPBB amounts of uranium.

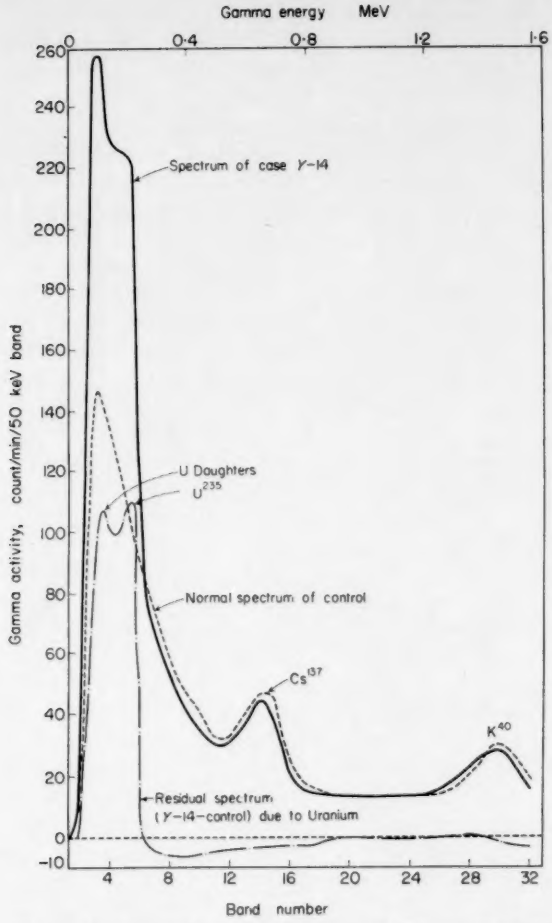

FIG. 6. Example of control-spectrum subtraction in an IVRM examination.

Although several isotopic abundance levels are considered in this and other sections and are indeed of interest, most of the experimental work was related to U^{235}-enriched or Level 1 materials and this paper gives primary attention to this isotopic abundance.

Estimation of normal in vivo *spectrum*

The *in vivo* gamma counting method proposes to determine uranium lung burden by measuring the gamma activity due to U^{235}, having an energy of 186 keV. The 186 keV region activity varies from person to person, even when no question of uranium exposure is involved, because of the different sizes of the thorax and variation in the body content of Cs^{137} and K^{40}. Although the pulse-height analyzer sorts the gamma radiation detected by the scintillator, incomplete absorption of the energy of a gamma quantum in the detector produces a finite number of counts in energy regions other than that of the maximum energy photopeak, even with as large a detector as the one in use. Regardless of the capture efficiency of the detector, a fraction of the radiation passing from the body undergoes scattering and is received as a lower energy quantum by the detector. It is to be expected that the scattering coefficients for the Cs^{137} and K^{40} in the body will be a function of body shape and size. Therefore,

the method must include some technique for estimating the normal count rate, not due to uranium, in the 186 keV region of the subject's spectrum.

For this reason, the *in vivo* spectrum taken in a routine chest examination includes not only the low-energy uranium region, but a full energy range to 1.6 MeV, in order that the data will reflect the photopeak and scattered radiation levels of all of the important gamma emitters in the body. In Fig. 6, a typical *in vivo* spectrum from a chest measurement is indicated as the "normal spectrum of control."

The most obvious technique of correcting for the normal 186 keV activity would appear to be the recording of the *in vivo* spectra of persons before possible uranium exposure. However, the factors of body size and Cs^{137} and K^{40} content are subject to change with time because of weight, diet, and fall-out of atmospheric contamination. These limitations necessitate the treatment of each IVRM examination independently of other examinations on the same subject at appreciably earlier times.

Control subject technique. The next obvious method for determining the normal count rate in the 186 keV region, and the method initially used in the IVRM program, is the selection of a control person, not subject to the radioactivity exposure hazard, who matches the subject in body shape and size and Cs^{137} and K^{40} content. Although body size and shape statistics can be easily accumulated, knowledge of Cs^{137} and K^{40} content requires an *in vivo* radioactivity measurement. In this program, a few persons not involved in uranium processing have been examined, including a number of newly hired employees. In general, however, the need for control subjects of a great variety of body sizes and Cs^{137} and K^{40} content has necessitated the use of other process workers whose spectra do not show obvious uranium spectra as controls. This procedure is recognized to carry the danger of underestimating lung burden because of possible low level of uranium lung burden carried by some of the control subjects.

Although the process of selecting a control subject for a questionable case involves consideration of all of the parameters of body size and normal *in vivo* radioactivity available, study of the data from subjects not considered to be obviously contaminated by uranium suggests that the most important factors involved are sex, body weight, and Cs^{137} content. Therefore, in selecting control subjects, primary attention is given to these factors. In order to evaluate the error involved in selecting controls, 21 pairs of controls, matching in weight and Cs^{137} content, were taken from the IVRM data. The weight range of these pairs was from 123 to 240 lb, and the Cs^{137} range from 122 to 210 counts/min. Statistical analysis indicated an agreement within individual matches of ± 8.6 lb in weight and ± 6.5 counts/min in Cs^{137} content (limit of error, x, 0.05)*. The agreement of 186 keV region (50 keV wide) between matches under these conditions was found to be ± 13.52 counts/min (limit of error, x, 0.05).

Fig. 6 illustrates the use of this technique. The control subject for Case Y-14 was chosen from the file of previous examinations on persons not previously carrying uranium. When the two spectra are subtracted, a residual due to the uranium lung burden of Case Y-14 is obtained. This example is typical, because the match of Cs^{137} and K^{40} can be noted to be less than perfect.

Spectral prediction technique. Early in the IVRM program, it was decided that an effort should be made to determine equations or systems for the calculation of the expected normal count rate in the uranium spectral region from knowledge of the body shape and size of the subject and the measured Cs^{137}, K^{40} and other naturally occurring activities. The requirement mentioned above, of a large population of persons not exposed to uranium inhalation or ingestion and on whom *in vivo* radioactivity measurements have been made, is again a limitation on this work.

The technique has been to use data on process workers whose spectra do not show obvious indications of uranium. The prediction equations derived were recognized to be subject to bias because of the possible low level of uranium contamination in the subjects.

* The notation "x, 0.05" here and with other statements of statistical analysis indicates the expected error or variation of a single measurement (x) at the 95 per cent confidence level (0.05).

The results of correlation studies of body size and *in vivo* radioactivity measurements are two equations, one for the prediction of the normal 186 keV spectral region, and another for the 90 keV region:

$$N_{186} = 12.1 + 0.7916\ (M_{290}) + 0.0708\ (K^{40}) + 0.107\ (Cs^{137}) + 0.096(W) \quad (1)$$

where N_{186} = predicted *in vivo* count rate in 165–215 keV band for normal *in vivo* chest spectrum, with no uranium contamination (counts/min); M_{290} = measured *in vivo* count rate in 265–315 keV band of *in vivo* spectrum in question (counts/min); K^{40} = measured count rate due to K^{40} photopeak in spectrum, 1.25–1.55 MeV (counts/min); Cs^{137} = measured count rate due to Cs^{137} photopeak in spectrum, 585–735 keV (counts/min); and W = body weight of subject (lb);
and

$$N_{90} = 7.0 + 1.144\ (M_{290}) + 0.2543\ (W) \quad (2)$$

where N_{90} = predicted *in vivo* count rate in 65–115 keV band for normal *in vivo* chest spectrum, with no uranium contamination (counts/min).

It is apparent that equations (1) and (2) are not equations for direct prediction of the scattered radiation intensity from various amounts of two radiation sources (K^{40} and Cs^{137}) in scattering structures with various scattering coefficients (human chests of various shapes and sizes). Rather, the equations use the measurement of scatter in one portion of the spectrum (265–315 keV, where 186 keV gamma quanta have essentially no effect) as the primary predictor for the scattered radiation intensity in lower-energy spectral areas (165–215 keV and 65–115 keV). The actual contributors of the scatter (Cs^{137} and K^{40}) and one of the parameters (body weight) which should determine the scattering coefficients are used as secondary predictors.

More direct correlations have been attempted, but none yield as accurate a prediction of the 186 keV area as equation (1). In the study, prediction equations were computed using combinations and multiples of the following variables: body weight, body height, chest thickness, chest width, Cs^{137} activity, K^{40} activity and the scattered radiation band activity (M_{290}). The failure to find a direct prediction of scatter suggests that the body size and shape parameters available for analysis are not sufficiently indicative of the scattering coefficients of the human chest.

The data used to derive equations (1) and (2) were primarily from men, but in testing with the limited number of women's spectra available, the equation seems to hold for either sex.

The estimated limit of error (x, 0.05) of predicting N_{186} by use of equation (1) is ± 11.4 counts/min (30 min counting period). The limit of error of predicting N_{90} by use of equation (2) is ± 22 counts/min.

The initial forms of equations (1) and (2) computed from 123 persons, mostly process workers, varied slightly in intercept from the form shown. The original equations were tested by predicting N_{186} and N_{90} for 21 newly hired employees who had not worked at an atomic energy installation prior to employment at the Y-12 Plant and on whom standard *in vivo* radioactivity measurements had been made. The measured count rates in the 186 and 90 keV bands fell within the limits of error of the predicted values, but the predicted count rates were found to have a significant positive bias (+3 counts/min for 186, +7 counts/min for 90 keV) in relation to the measured activity. These biases indicate that some of the 123 persons used in computing the equation actually contained low levels of uranium contamination, as was suspected. The final forms of the equations have been corrected for this bias.

The value of such prediction equations is substantial, particularly for an entirely routine, large scale IVRM program. Use of such equations eliminates the need of control selection. The equations also eliminate the attendant source of error in using a control when the desired match is not available in the file of control subjects. The present equation for the 186 keV region is comparable to control matching under the best of matching conditions available.

It is proposed to continue to collect IVRM data on persons not exposed to uranium and to continue efforts for more accurate prediction equations from these data.

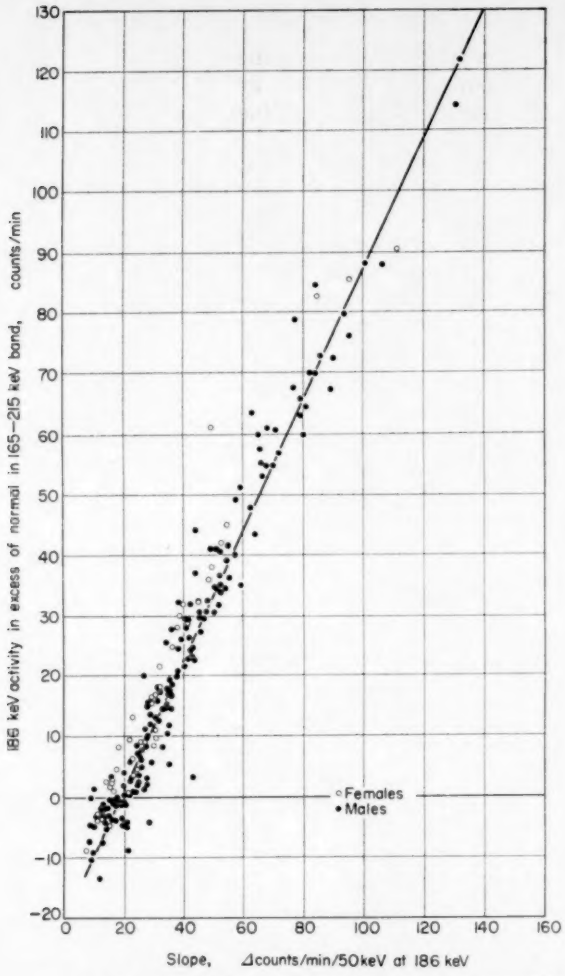

FIG. 7. Relationship of spectral slope to uranium activity

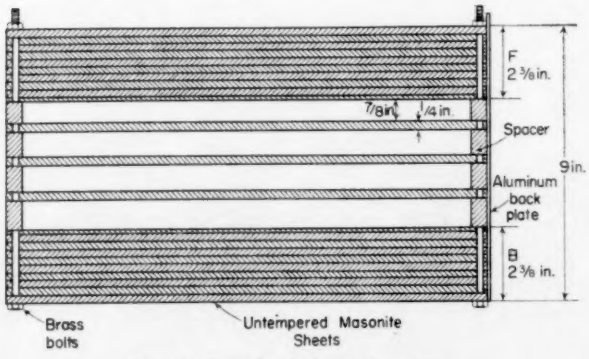

FIG. 8. Standard chest phantom.

The prediction equation technique is now used on routine *in vivo* radioactivity measurements for uranium. Selection of control subjects is still used upon occasion for special cases.

Spectral slope technique. In the course of the work, another approach to the same basic problem was developed. When the lung burden is high enough, no difficulty is encountered in concluding even from the analyzer's cathode ray tube display of the gross *in vivo* spectrum, that uranium is present. In cases of one MPBB or less, judgment from the gross spectra itself is more difficult. In these cases, the most reliable indication of the presence of uranium has been found to be the slope of the gamma spectrum at the 186 keV region. Although the Cs^{137} content and body size cause large variations in the count rate of the entire low-energy region, the slope of the spectrum, taken as the difference between the count rates in the 165–215 keV and the 215–265 keV bands, remains reasonably constant unless uranium is present. During the development of the method, it became customary to regard spectra having a slope of greater than 30 as requiring close attention.

Subsequently, when the more refined method of the prediction equations had been developed, the count rate in excess of normal in the 186 keV region was determined for a large number of examinations and plotted as a function of slope, as shown in Fig. 7. It is interesting that the residual activity equivalence of the slope value of 30 is about 12 counts/min, essentially the upper limit of the error involved in using equation (1). Thus, the slope concept can be used for a very rapid indication of the presence of uranium-235 in the subject spectrum.

Relation of uranium in vivo *activity to uranium lung content*

Having estimated the normal spectral activity in the uranium regions, the method must then provide a calibration relating the excess activity to uranium lung content.

The basic calibration method has been the measurement of standards in an untempered Masonite chest phantom of the type devised by MILLER at Argonne National Laboratory for the initial measurements on Case Y-1. As described by MILLER[1], the technique used in designing this simulation of the human chest was the adjustment of the front and back wall thicknesses until the ratio of 90 keV/186 keV gamma lines of standards placed in the phantom was the same as that of the residual *in vivo* spectrum of Case Y-1.

A view of this phantom, which is about 15 in. × 15 in. in height and width, is shown in Fig. 8. The thick sections at the top and bottom (F and B) simulate the body wall, front and back, and the open area simulates the cavity or low-density volumes of the chest.

When standards are placed at various positions within a 6-in. diameter circle in the midplane of the phantom and the average count rate of the given amount of uranium is computed under the conditions of uniform distribution over the circle, the distributed 186 keV count rate is found to be 86 per cent and the 90 keV count rate 82 per cent of the count rates from the centered standard. When these geometry factors are applied to the centered count rates for standards of the three levels of uranium enrichment, the calibration data of Table 2 are obtained.

The variations in the U^{235} per counts/min as the

Table 2. Uranium calibration factors from standard chest phantom

Type factor	Level 1 uranium	Level 2 uranium		Level 3 uranium	
	186 keV	186 keV	90 keV	186 keV	90 keV
μg U per counts/min*	10	935	4440	1613	3840
μg U^{235} per counts/min*	9.3	6.5		3.6	

* Activity in excess of normal in 65–115 keV band for 90 keV factors; in 165–215 keV band for 186 keV factors.

enrichment level changes may be understood by reference to Fig. 5, where it will be noted that the large ratio of 90 keV/186 keV peak heights for Levels 1 and 2 uranium causes measurements of the 186 keV activity to include an appreciable contribution from the "tail" of the 90 keV peak. Although this factor causes an increase in sensitivity to U^{235} on the basis of 186 keV counts/min per μg for the lower U^{235}-enrichment levels, it also means that for these enrichments the 186 keV activity measurement will be affected by any uncertainty regarding the equilibrium state of the uranium daughters.

The method of designing a phantom and making a calibration for each individual examination is not practical in a routine program, because of the time involved and also because few cases have sufficient lung burden to produce a clearly defined residual spectrum to which the phantom spectrum can be adjusted. So far, the calibrations from a single standard phantom (Table 2) have been used for all individuals examined. At the same time, efforts are underway to evaluate the error because of different body sizes of the individuals and to develop more refined techniques of achieving individual calibrations which are also amenable to routine use.

In the first place, phantom adjustments and calibrations can be made on any cases having large, clearly defined residual spectra in order to establish a range of the calibration factors over which correction should be made. Secondly, data is being collected regarding the absorption by the full body thickness of uranium gamma quanta from samples placed beneath the subject. The assumption is made here that the front and back walls of the body have equal effects in absorbing and scattering the radiation and that the absorption data for transmission through the full body thickness can be extrapolated to a linear absorption coefficient for uranium placed in the midplane of the upper thorax. In such a consideration, each inch of chest thickness is treated as a $\frac{1}{2}$ in. of absorber addition in the front wall. The preliminary data on this work suggests that the full thickness of the human chest exhibits a linear absorption coefficient of about 0.169 cm^{-1} for 186 keV radiation. This value is less than the 0.199 cm^{-1} measured for solid untempered Masonite in the chest phantom and larger than the 0.142 cm^{-1} to be expected of water, the usual equivalence of soft tissue.

Table 3. Calibration correction factors for chest thicknesses

Chest thickness (in.)	Calibration correction factor*
7.0	1.19
7.5	1.10
8.0	1.00
8.5	0.90
9.0	0.81
9.5	0.73
10.0	0.65

* To be divided into the 186 keV calibration factors of Table 2.

The chest thickness of Case Y-1, on which the standard phantom is based, is about 8 in. If the linear absorption coefficient discussed above is used with a factor of 0.5 in. absorber thickness for each 1 in. of total chest thickness, correction factors to be applied to the 186 keV calibration data of Table 2 may be computed. Such factors are shown in Table 3.

These factors, and indeed this approach to the problem, are still under evaluation and are only reported to indicate the direction of present approaches to the solution of the problem of varying chest thickness.

The need for such corrections may be deduced from Table 4, in which data regarding the chest thickness variations observed in a large number of routine examinations are presented.

RESULTS

Analysis of routine examinations

The development program of routine *in vivo* radioactivity measurements on some Y-12 employees working in uranium process areas is summarized in Table 5. The scheduling for this program was planned to examine persons whose work categories and/or work areas suggested that they might be exposed to inhalation of uranium. The persons selected for examination are classified in the table into three

Table 4. Chest thickness variations observed

Category	Examinations	Chest thickness (in.)			
		Average	Minimum	Maximum	Range of 67% of examinations
All examinations	535	8.37	6.3	11.8	7.4–9.3
Male examinations	474	8.46	6.3	11.8	7.5–9.4

Table 5. Summary of routine in vivo *radioactivity measurements*

Group	Group description	No. in group	No. of cases in which detectable uranium spectra were observed	%
A	Lowest probability of exposure	65	4	6.2
B	Medium probability of exposure	111	17	15.3
C	Highest probability of exposure	65	28	43.1

Table 6. Case Y-1—uranium inhalation

Date	Lung burden estimates		Uranium excreted between measurements	
	IVRM Uranium† (mg)	Urinalysis Uranium (mg)	IVRM Uranium (mg)	Urinalysis Uranium (mg)
1 November 1957	2.75*	2.72	—	—
19 November 1957	2.05*	1.37	0.70	0.74
10 December 1957	1.47*	1.00	0.58	0.48
18 March 1958	0.68*	0.52	0.75	0.78
13 June 1958	0.33	0.31	0.35	0.22
21 August 1958	0.27	0.21	0.06	0.09
8 October 1958	0.21	0.23	0.06	0.02
2 April 1959	0.11	0.05	0.10	0.06

* Measurements made by Dr. C. E. MILLER at Argonne National Laboratory.
† Uranium of Level 1 U^{235} concentration, Table 1.

groups according to probability of exposure.

In cases in which the slightest indication of uranium lung burden has been detected, re-examinations have been scheduled and attempts made to continue the observations until the *in vivo* spectrum appears normal.

Example results

The best example of the results of *in vivo* uranium measurements is the Case Y-1. This employee was the subject of the initial *in vivo* uranium measurement made at Argonne National Laboratory[1,2] as described in the Introduction. The measurements were continued in the Y-12 IVRM facility. The case data is particularly significant because the employee was removed from the uranium process areas after the initial exposure and repeatedly measured over a rather long period of time. In addition, urinalyses for uranium,

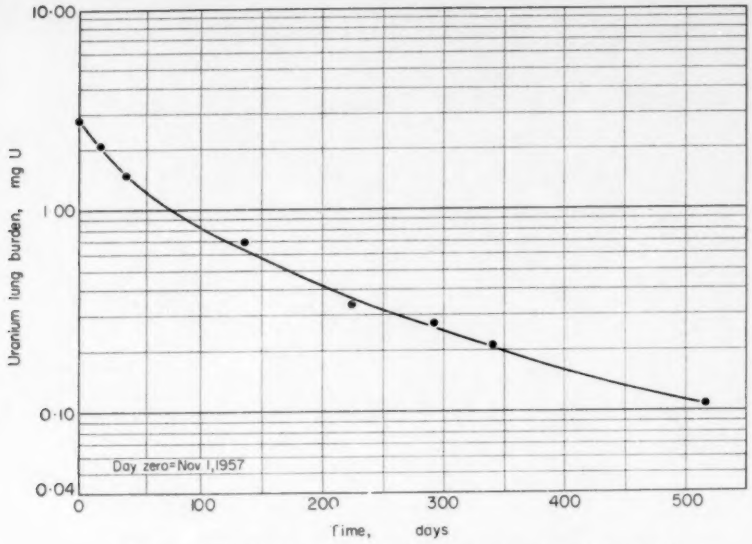

FIG. 9. Elimination of a uranium lung burden—Case Y-1.

made over the period of the *in vivo* radioactivity measurements, were available for comparative studies of the two methods. The urinalysis and IVRM data on this case are summarized in Table 6.

Fig. 9 is a graph of the uranium lung burden of Case Y-1 at various times when IVRM data was taken. The initial biological half-life seemed to be 40 days, and a latter half-life about 120–150 days.

All aspects of this case have been examined and reported in detail by the Y-12 Health Physics Department.[9]

Table 7 represents the *in vivo* gamma counting measurements on three other cases, over shorter periods of time.

Table 7. *Data on uranium lung burden measurements by* in vivo *gamma counting*

Case No.	Date	Estimated lung burden of Level 1 uranium	
		Uranium (μg)	MPBB (%)
Y-14	24 September 1958	1072	390.4
	8 October 1958	550	200.2
	3 December 1958	289	105.2
Y-24	14 November 1958	925	336.8
	26 November 1958	872	317.6
	5 December 1958	489	178.1
	12 December 1958	244	88.9
Y-25	19 January 1959	800	291.3
	26 January 1959	699	254.6
	2 February 1959	673	245.1
	11 February 1959	550	200.3
	18 February 1959	548	199.6

The measurements on Case Y-14, one of which was used in Fig. 6, may be used as examples of the graphical appearance of the *in vivo* gamma spectrum when uranium is present. Fig. 10 is the net *in vivo* gamma spectrum of Case Y-14 at each measurement. A single line represents the three spectra above 250 keV, which are identical within the counting statistics. The relative heights of the uranium region (90 and 186 keV) can be observed for each measurement. In Fig. 11, the *in vivo* spectrum of a control subject matching Case Y-14 in Cs^{137}, K^{40} and body size has been subtracted from each spectrum of Case Y-14, leaving the residual *in vivo* gamma spectrum of uranium. Finally, the same correction procedure has been performed in Fig. 12 on a chanel-by-channel basis to give better resolution of the uranium lines.

EVALUATION OF METHOD

Statistical evaluation

Considerable statistical evaluation has already been made in the discussions of the techniques of the method. In addition, the ability of the *in vivo* radioactivity measurement to reproduce the *in vivo* spectrum has been determined by repeated counting runs on the same person in a single day. The statistical analysis indicated that the variance between measurements of the 186 keV band, the K^{40} photopeak and the Cs^{137} photopeak in separate examinations is not

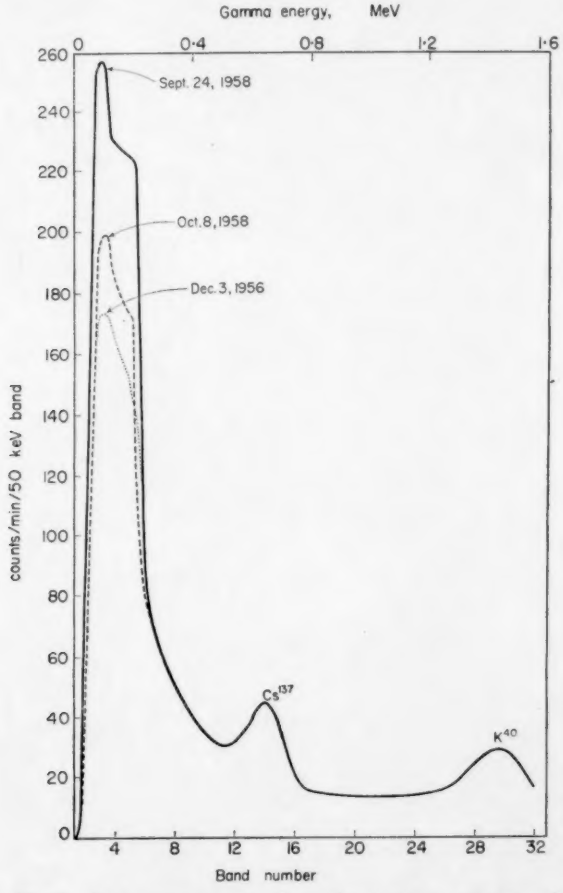

FIG. 10. *In vivo* gamma spectra from three measurements—Case Y-14.

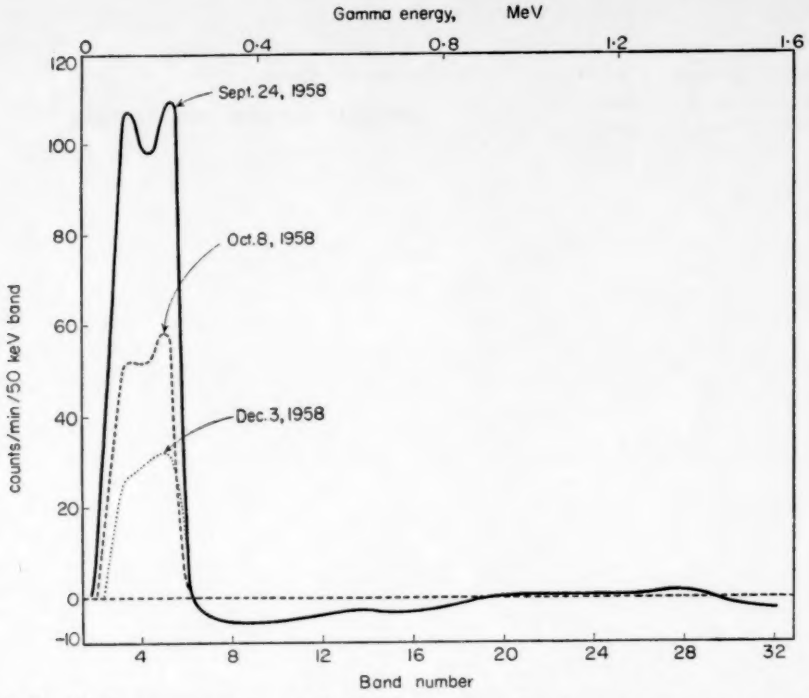

FIG. 11. Residual gamma spectra attributed to uranium for three measurements on Case Y-14—corrected for normal *in vivo* gamma activities by control-spectrum subtraction technique.

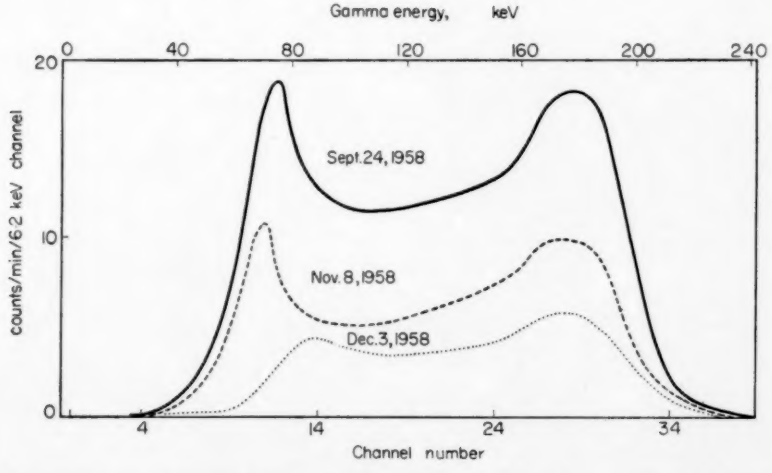

FIG. 12. Residual gamma spectra attributed to uranium—high resolution spectral graph for Case Y-14.

Table 8. Summary of statistical evaluation

Parameter number	Description of parameter	Assumed value (typical) of 186 keV band, (counts/min)	Count time (min)	Limit of error for single measurement, 95% confidence level (counts/min)
1	Gross count rate (subject carrying equivalent of 1 MPBB of Level 1 uranium)	237	10	±9.6
		237	20	6.7
		237	30	5.5
2	Background count rate	90	50	2.6
3	Net *in vivo* count rate, (1)–(2)	147	10	9.9
		147	20	7.2
		147	30	6.1
4	Predicted normal count rate, (equation 1)	120	30	11.1
5	Net U^{235} count rate, (3)–(4)	27	30	13.1
6	Theoretical control subject match, (3) minus perfect control	27	30	8.4
7	Experimentally measured error of control machining	0	30	13.5
8	Reproducibility of the measurement, (net *in vivo* count rate, repeated on same subject)	147	30	6.1
9	Uranium lung burden, from (5) or (7), using factors of Table 2			
	Level 1 uranium*		30	±131 μg U = 47.6% MPBB*
	Level 2 uranium*		30	12,248 μg U = 47.8% MPBB*
	Level 3 uranium*		30	21,138 μg U = 42.6% MPBB*

* As described in Table 1.

significantly different from the counting variance.

Table 8 summarizes this and other parameters considered in determining the accuracy and precision of the *in vivo* gamma counting method of measuring uranium lung burden and on which limits of error can be assigned at present. Some parameters on which error contribution evaluation has not been completed include surface and hair contamination, clothing activity, and body wall absorption differences.

Limitations of the method

I. A single *in vivo* radioactivity measurement cannot determine if the uranium detected is from inhalation of soluble or insoluble uranium compounds.

II. It is desirable to know the level of uranium to which a person has been exposed, although the relative height of 90 and 186 keV lines is an approximate indicator of enrichment level.

III. Surface contamination is a possible source of error in the method, even when careful showering is required. Frequently, 186 keV activity has been measured from the hair of the subject after showering. The hair is perhaps the most probable carrier of surface contamination. Fortunately, the detector shield used in the Y-12 IVRM facility is of great help in eliminating the interference of uranium hair contamination. With 100 mg of Level 1 uranium at the crown of the head, no contribution is added to the 186 keV region in the

standard chest measurement. In the case of women, where the hair is longer and the head may not be between most of the contamination and the detector, the shielding is evidently not sufficient to eliminate the effect totally. Occasionally, particularly with subjects who have large amounts of chest hair or who have not showered thoroughly, surface contamination has been discovered on the chest. Such contamination can provide a large positive bias in the apparent lung burden, since the calibration factor for uranium in contact with the crystal face is about ten times as large as that for uranium in the lung. An alpha survey meter is used to check the chest surface to reveal these cases. Also, the occurrence of surface uranium contamination can usually be recognized by the ratio of 90 keV/186 keV peaks, which is lower for surface activities than for internal uranium.

IV. Additional work is required and is underway to evaluate the error involved in using a single calibration factor for all persons, regardless of chest thickness, and to devise more accurate quantitative calculation methods.

V. More accurate methods of predicting the normal 186 keV count rate of a given subject would be desirable and are being sought. Doubtlessly, the availability of IVRM data on a large population of persons free of possible uranium contamination would improve the accuracy of this part of the procedure. Ultimately, it would be desirable to reduce the limit of error on Parameter 5 of Table 8 to the level of Parameter 6.

VI. The capacity of the single IVRM facility is relatively small compared with the current urinalysis program of periodic monitoring for inhalation exposure. Additional work is necessary to determine the required frequency of examination by the IVRM method, the minimum examination time required, and the possibilities of more rapid processing of IVRM data. The latter problem is being considered in terms of recording the counting data in a format or medium suitable for automatic computation, using the spectral prediction equations.

CONCLUSIONS

In the development of the *in vivo* radioactivity measurement program, it has been shown that quantities less than the maximum permissible body burden amounts of insoluble uranium in the human lung can be detected and measured by gamma scintillation spectrometry. Although several aspects of the problem need additional development, the method is already useful as a health physics tool. In routine use for monitoring personnel possibly exposed to uranium enriched to 93 per cent in U^{235}, the *in vivo* method is easily capable of detecting the presence of 130 μg of U^{235}, equivalent to 50 per cent of the MPBB of insoluble enriched uranium, in the human lung. The calibration factors and measurement limits of error computed indicate that measurements of as low as 50 per cent MPBB can be detected for any level of U^{235} enrichment considered. Quantitative interpretations of the uranium activities detected by the IVRM method require some knowledge of the U^{235} enrichment level of the uranium in question, although the relative heights of the 90 and 186 keV peaks in the *in vivo* spectrum is an indicator of U^{235} enrichment.

The method includes techniques for estimating the normal *in vivo* spectrum in the uranium gamma energy regions and for relating the uranium *in vivo* count rate to uranium lung content.

As a measurement tool for known exposure cases, greater precision and lower limits of detection could be attained in periodic measurements of the gradually diminishing amount of uranium present in the lungs.

The ability to measure as low as 50 per cent of MPBB of insoluble uranium in the human lung, without dependence upon excretion analysis, makes the *in vivo* gamma counting method a valuable adjunct to the health physics program concerned with processes in which uranium inhalation is possible.

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URINALYSIS METHOD FOR ENRICHED URANIUM

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Abstract—A urinalysis procedure was developed for the estimation of micro-amounts of enriched uranium which are coprecipitated with an ammoniacal alkaline earth phosphate precipitate. The precipitate is fired in a muffle furnace and the enriched uranium purified by anion exchange. The isolated enriched uranium is electroplated and determined by autoradiography. The method has a sensitivity of 0.015 ± 0.007 d/m per 150 ml of sample, a recovery of 94 per cent, and a standard deviation of ±16 per cent.

INTRODUCTION

THE analysis of urine samples is the principal method used to monitor personnel for the possible assimilation of many radionuclides. A urinalysis method must be easily adapted to a routine bio-assay program in which large numbers of personnel may be sampled. Due to the harmful effects of very small amounts of radioactivity located in certain parts of the body, the analytical method must be sensitive enough to detect extremely small quantities of the radionuclides. A minimum sensitivity of 7.6 d/m per 24-hr urine specimen (1.5 liters) is necessary to detect one-tenth the suggested tolerance level of enriched uranium in the body.[1]

The fluorimetric method is adequate for natural uranium urinalyses but lacks the sensitivity necessary to detect biologically significant concentrations of enriched uranium (containing approximately 1% U^{235}). The method employing electrodeposition described in this paper will detect micro-amounts of enriched uranium in 150 ml samples of urine.

PROCEDURE

Reagents. All chemicals used are of reagent grade and no further purification is necessary. The calcium carrier solution is made by dissolving 111 g calcium nitrate, $Ca(NO_3)_2.4H_2O$, in 100 ml of distilled water.

Ion exchange column. The column, a 10 cm × 6 mm diameter tube with a 30-ml reservoir bulb at the top, is packed to a 10-cm height with a 0.1 N HCl slurry of Dowex 1 × 10 (100 to 200 mesh) anion exchange resin. The resin is previously cleaned and sized by washing with several portions of 0.1 N HCl. The resin bed is prepared by passing 5 ml of 8 N HCl through the column.

Electrodeposition apparatus. A Fisher Electro-Analyzer equipped with hot plates is used. The electrodeposition cell consisted of a "Lucite" cylinder with a screw-on stainless steel cap containing a shimplate, and a ½-in. stainless steel plating disc.[2] Uranium is deposited on a circular area 6 mm in diameter.

Sample preparation. To a 150-ml urine sample in a 1-liter beaker, add 5 ml of concentrated nitric acid, 2 ml of 85.8% ortho-phosphoric acid, and dilute to 500 ml with distilled water. Add 1 ml of the calcium carrier solution, and heat to 85°C. With stirring, slowly add concentrated ammonium hydroxide until precipitation occurs, and then add 10 ml in excess. Continue to stir and heat the solution at 85°C for 2 hr. Cover the beaker with a watch glass and let the precipitate settle overnight. Decant the supernate, being careful not to disturb the precipitate. Using a Fisher Filtrator funnel and a suction flask, quantitatively transfer the precipitate to a Whatman No. 50 filter paper. Place the filter in a 30 ml Vycor crucible, and place the crucible in a cold muffle furnace. Fire the precipitate at 900°C until it is white; cool, and dissolve the residue in 10 ml of 8 N hydrochloric acid.

Ion exchange. Quantitatively transfer the solution to a previously prepared anion exchange column. After the solution has passed through the column by gravity feed (flow rate 0.25 ml/min), wash twice with 10 ml portions of 8 N hydrochloric acid. Elute the enriched uranium by passing 15 ml of 1 N hydrochloric acid through the column.

Electrodeposition. After evaporating the elutriant

from the column to dryness, dissolve the residue in 0.5 ml of nitric acid and 3 ml of saturated ammonium oxalate. Quantitatively transfer the solution to a previously prepared electrodeposition cell with two additional 3 ml portions of saturated ammonium oxalate. Immerse the cell in a water bath maintained at from 85° to 90°C. Electroplate for 2 hr with stirring, at a current of 300 mA. Maintain the liquid level in the cell during the electrolysis by the addition of saturated ammonium oxalate.

Make the electrolyte basic by the addition of 2 ml of 6 N ammonium hydroxide and electroplate for 3 hr more. Color indicators tend to interfere with plating. Basicity and liquid level are maintained by the periodic addition of 6 N ammonium hydroxide to keep the cell full until electrodeposition is complete. Remove the cell from the electrodeposition apparatus, and pour off the electrolyte as quickly as possible. Rinse the cell with distilled water and remove the stainless steel disc. Dry the disc thoroughly under infrared light. Count the disc for 10 min in an α-counter. If the count exceeds the counting sensitivity, obtain an accurate rate by further counting.

Radioautographing. If the count is less than counter sensitivity, radioautograph the plate for 1 week using Kodak NTA emulsion plates mounted in a brass camera.[3] Eradicate tracks from the NTA emulsion slides before exposing them to the plated discs.[4] Process the NTA slides using Kodak D-19 developer and Kodak F-5 fixer solution.[5]

Track counting. Use a Leitz Microprojector, model Xl-C, to count all tracks which are within a 12.92 mm^2 area of the emulsion.[6] Make a background count on an unexposed portion of the film, and subtract it from the sample count. Calculate the amount of enriched uranium by comparing the number of tracks from the sample with the average number of tracks found in the analysis of a large number of urine samples spiked with a known amount of enriched uranium.

DISCUSSION

The ammoniacal phosphate precipitation utilizes calcium carrier added to the urine to form a general scavenging precipitate. The solution is heated and stirred for 2 hr to get complete coprecipitation and coagulation of the precipitate. The precipitate is fired to eliminate organic matter before treatment with the anion exchange resin.

The absorption behavior of uranium-(VI) on Dowex 1 anion exchange resin has been studied in HCl solutions.[7] The absorbability of uranium increases rapidly with increasing HCl concentration to a peak absorption near 8 M HCl. At higher concentrations the absorbability decreases slightly. Since the absorbability of uranium on the resin is negligible at low concentrations of hydrochloric acid, the metal is easily eluted with a minimum volume of 1 N HCl.

The evaporation of the elutriant to dryness is necessary to obtain the enriched uranium in the desired electrolyte. The higher specific activity of enriched uranium makes the electrodeposition from common electrolytes at low-activity levels more difficult than natural uranium.[8] However, by plating for 2 hr in a saturated ammonium oxalate electrolyte made slightly acid with 6 N nitric acid and then for 3 hr in the electrolyte made basic by the addition of 6 N ammonium hydroxide, quantitative electrodeposition of submicrogram amounts of enriched uranium is obtained. A temperature of 85°C and continuous stirring are also necessary to obtain high recoveries.[9] The optimum plating time is 5 hr.

In order to obtain a faster (2 vs. 7 days) analysis, the electrodeposition step can be eliminated and the sample directly plancheted and counted following the ion exchange step. The uranium isolated from all extraneous material by the ion exchange step makes for a clean planchet, free of any residue. However, by increasing the speed of the analysis in this way the sensitivity is limited by the counting method. This lowered sensitivity (about 100-fold less sensitive) can be somewhat overcome by increasing the amount of sample in order to obtain larger amounts of enriched uranium. With samples over 500 ml no calcium carrier is needed as there are sufficient alkaline earths present in the urine to form the scavenging precipitate (1 ml of calcium carrier solution is sufficient for samples up to 500 ml volume).

A pulse-height analysis of the plated disc or plancheted sample can be used to determine the uranium isotope isolated.

RESULTS

An average recovery of 94 per cent and a standard deviation of ± 16 per cent was obtained on a series of one d/m enriched uranium spikes. A sensitivity limit of 0.15 ± 0.07 d/m per 1.5 liters of urine was achieved using a 150-ml urine

specimen and an exposure time of the plated disc to the NTA slide of 1 week.

Less than 13 per cent of any plutonium, and less than 0.1 per cent of any thorium contaminating the specimen are recovered by the method.

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ELECTRODEPOSITION OF URANIUM FROM URINE

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Abstract—Electrodeposition of uranium from wet ashed urine solutions shows a wide variation of recovery between different specimens. This lack of precision is generally attributed to a co-deposition of inorganic salts which reduces the detectable alpha activity by absorption. A series of samples was plated after adding to aqueous solutions of uranyl nitrate the major cations normally found in urine—calcium, magnesium, sodium and potassium. The principal trouble-causing elements are calcium and magnesium, and their removal as the carbonate and the phosphate, respectively, was effective in eliminating the extraneous salt deposits on the plates. Recoveries from 33 different urines tested averaged 89.5 ± 4 per cent.

INTRODUCTION

THE electrolytic deposition of uranium on metal foil has been recognized for a number of years as an efficient method for the isolation of the element from an aqueous solution of its salts. COHEN and HULL[1] established a quantitative method for the plating of uranium from solutions of uranyl salts and measured the deposited uranium by alpha counting. ROTHSTEIN[2] *et al.* modified some of the established methods and were successful in determining the uranium content of biological materials by first isolating the uranium by precipitation and/or ether extraction and then plating on silver foil. WHITSON and KWASNOSKI[3] published a method for the determination of uranium alpha activity in urine which involves prior precipitation of calcium as the oxalate.

The Applied Radiobiology Section bioassay laboratory had for several years used a modification of the COHEN and HULL method for electroplating and had been successful in assaying a large number of urine samples with reasonable precision. The urine samples were digested in nitric acid and transferred directly to the plating cell. All of the inorganic salts normally found in urine were present in the cells and precipitated during plating. Many of the discs were covered with a deposit of these salts which leads to a considerable reduction in alpha counting efficiency because of absorption Furthermore, some of the discs showed non-uniform flaking of the deposit which resulted in variable loss of material and markedly increased the possibility of contaminating the low background alpha counters. In order to make maximum use of existing equipment, the electroplating method was investigated to determine the degree to which recovery and reproducibility could be improved.

EXPERIMENTAL

Electrodeposition unit

The cell components and their order of assembly are shown in Fig. 1. An ordinary 4-oz glass bottle with the bottom removed forms the chimney. The silver foil, on which the uranium is to be deposited, is placed between the brass cathode and an annular rubber gasket. A metal cap seals the cathode, silver foil and gasket to the glass chimney. The cathode lead extends through a center hole in the cap. A lucite cell plug serves to support the anode and thermometer and is loosely fitted to the glass chimney to allow for thermal expansion.

The anode is a flat spiral of 0.099 in. diameter platinum wire. This design provides for essentially constant current density between the electrodes and permits free circulation of the electrolyte.

Components of the cell are mounted on a

* Operated by Union Carbide Nuclear Corporation for the United States Atomic Energy Commission.

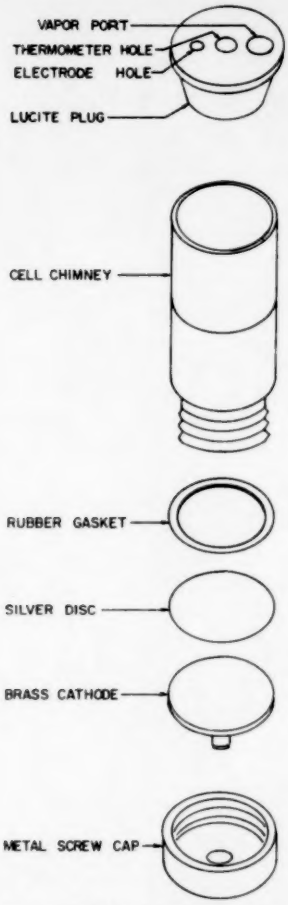

FIG. 1. Plating-cell assembly

shelf on the front of the power supply (Fig. 2). The anode is connected to the power supply by means of an insulated clip lead. The cell is tilted 20° from horizontal to improve the natural circulation throughout the solution, thereby eliminating the need for an auxiliary stirrer.

The heating jacket which is placed around the chimney consists of a $2\frac{1}{8}$-in. diameter copper sleeve 2 in. in length wrapped with an insulated heating element. Temperature is controlled by means of a variac.

DISCUSSION

In the development of the method it was found that trace amounts of uranium could be plated from a pure aqueous solution with a recovery of 92.3 $\pm$ 3 per cent. It was noted also that this high recovery could be obtained from some urine solutions while others gave recoveries as low as 20 per cent. By plating a large number of urine samples in which the uranium content was known, it appeared that the recovery rate for uranium was inversely proportional to the specific gravity of the urine specimen (Fig. 3). One possible explanation for this effect was the fact that the electroplating cell being used had no provision for externally heating the sample and the plating temperature (95–97°C) had to be maintained by adjusting the current flow through the bath. Consequently, since the total salt concentration was higher in the case of high specific gravity urines, a greater current was required to maintain that temperature. The higher currents seemed to cause a greater co-deposition of unwanted salts. By providing external heat to the cell with a heating jacket, current density curves were run on several urines of varying salt concentrations. It was found that the per cent recovery of low specific gravity urines showed little or no change after a saturation current was reached. The high specific gravity urines, however, showed a decreased alpha count rate on the plate after a threshold current was reached (Fig. 4). These plates also showed increased deposition of unwanted salts at the higher current. This seemed to indicate that, while uranium could be plated over a wide range of current density with little variation in recovery, the unwanted salts showed increased deposition as current was increased. Hence, the apparent recovery of uranium would be lower owing to absorption of the alpha particles by the extraneous deposits.

In an effort to determine the composition of these undesirable deposits a series of samples was plated after adding the major cations normally found in urine to aqueous solutions of uranyl nitrate. The cations—calcium, magnesium, sodium and potassium—were tested separately to determine the effect of each. The samples to which calcium was added showed immediate precipitation of the oxalate when mixed with the buffer solution. After plating, the discs showed some calcium deposited, although part of the precipitate remained in suspension in the plating bath. The measured

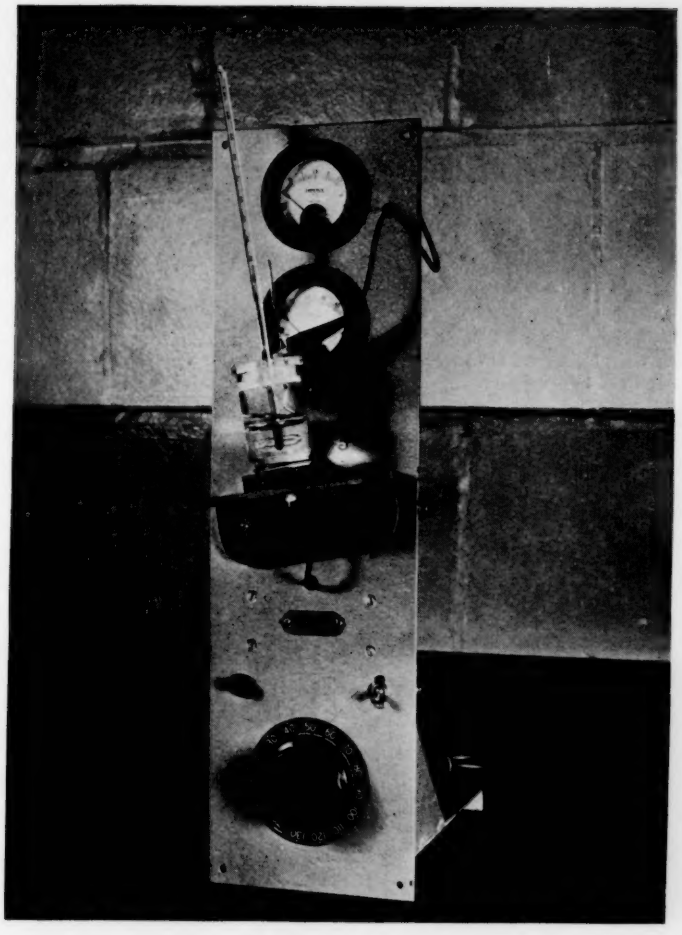

FIG. 2. Power supply

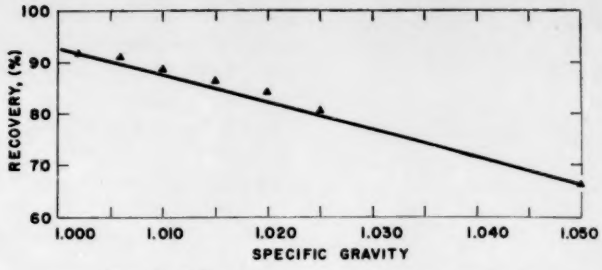

FIG. 3. Effect of specific gravity on recovery

alpha activity on the disc was lowered owing to absorption and possibly owing to adsorption of the uranium onto the particles remaining in suspension and discarded. Samples to which magnesium was added showed no immediate precipitation, but as the pH of the plating bath increased owing to decomposition of the buffer a precipitate formed which again was partially deposited, and part remained in suspension. The uranium recovery was also low. Samples to which potassium and sodium were added showed no precipitation and no deposition. The uranium recovery was the same as that of the controls to which no extra cations were added. Recovery of uranium from this series of samples is shown in Fig. 5. Sodium carbonate, which is one of the stronger complexers for uranium, was added to urine solutions to hold the uranium in solution while removing the calcium as the insoluble carbonate. Magnesium is removed as the phosphate. Removal of calcium and magnesium from the digested urine solutions was very effective in eliminating the extraneous salt deposits on the films. Recoveries from 33 different urines averaged 89.5 ± 4 per cent.

Because of the high uranium content of urine analyzed at this laboratory, not more than 25 ml of urine is needed for reasonable counting statistics. It is felt, however, that with adequate pretreatment, larger amounts of urine may be processed when the uranium content is very low.

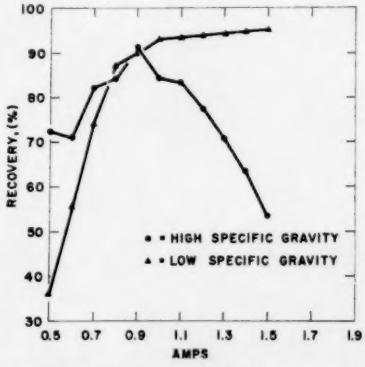

FIG. 4. Effect of current density on urines of different salt concentrations.

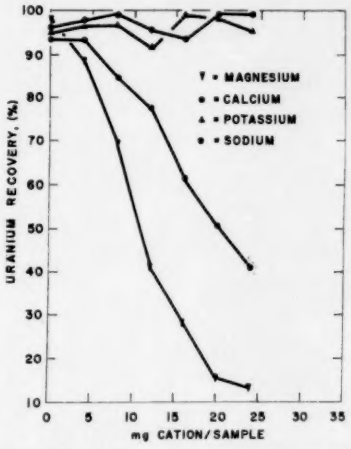

FIG. 5. Effect of major cations normally found in urine on electrodeposition of uranium

ANALYTICAL PROCEDURE

Buffer solution—(mix in 1 liter)
29 g ammonium oxalate
25 g sodium phosphate (tribasic)
0.218 g ferric ammonium sulfate
1 g bromecresol green

Concentrated HNO_3 is added to a fresh urine sample until all salts go into solution. Aliquots

of the HNO_3–urine mixture representing 25 ml of urine are digested on a steam bath with repeated additions of HNO_3 until the residue is soluble in 0.1 N HNO_3. After digestion the ash is dissolved in 10–15 ml 0.1 N HNO_3 and transferred to a 50 ml centrifuge tube; 5 ml 10% Na_2CO_3 and 1 ml 20% $NH_4H_2PO_4$ are added and the sample is heated for 1 hr in a steam bath. This treatment effectively removes calcium as the carbonate, and removes most of the magnesium. The carbonate forms a strong soluble complex with uranium which remains in solution. The samples are centrifuged 15 min at 1500 rev/min and decanted into the assembled plating cells. The precipitate is washed twice with a 1% Na_2CO_3 solution. HNO_3 is carefully added to the plating cell by drops until a pH of 3 is obtained. A 20 ml portion of the buffer solution is added and the pH is adjusted with 0.1 N HNO_3 to 4.5–5. The cells are then mounted on the plating unit and the current is adjusted to 2.0 A. The current will increase as the solution heats and is adjusted to 1.0 A when the temperature reaches 95–97°C. Plating time is 1 hr, during which period the temperature is maintained at 95–97°C and the current held constant at 1 A. After plating, the cell is decanted, rinsed with distilled water, disassembled and the film dried prior to counting.

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POWER FUNCTIONS RELATING EXCRETION TO BODY BURDEN

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Abstract—Formulae necessary to relate the quantity of radionuclides excreted to that assimilated in exposures that are acute and those that are multiple or continuous are derived from power-function relationships. Particular attention is given to providing equations having variables for which the bio-assayer can easily derive numerical values.

INTRODUCTION

EVIDENCE accumulated since 1946 indicates that the decrease in body retention of many bone-seeking elements, when administered as soluble compounds readily accessible to circulating fluids, may be better described in terms of a power rather than an exponential function. In laboratory animals, this was found to be true for calcium,[1] strontium,[2–5] and for radium.[1,6–9] In humans, NORRIS *et al.*[7] have shown that all available data from studies of radium metabolism over a 25-year period following administration can be better fitted by power functions. STEHNEY and LUCAS[10] used the power function to describe the increase in the radium burden of human subjects at the natural level of dietary intake. LANGHAM *et al.* presented data to show that retention and excretion of plutonium by man over a 5-year period can also be described in this manner.[11–13]

A case in which soluble plutonium was accidentally assimilated through a wound by a normal, healthy individual at the Savannah River Plant has been followed for over 4 years.[14] Here the decrease in the urinary excretion rate could also be best described in terms of a power function. Similarly, BERNARD and STRUXNESS[15] have found it applicable to the description of uranium retention by humans.

Admittedly, the application of the power function to the description of the metabolism of bone-seeking elements is based on experimental observations and is of little value in explaining the mechanisms involved. However, it is able to describe in simple mathematical fashion the fraction of certain radionuclides excreted per day, over extended periods of time. For this reason the revised report of the Committee on Permissible Dose for Internal Radiation[16] includes an appendix on the calculation of the maximum permissible concentration in air and water based on a power-function model.

Now bio-assayers are expected to interpret the dose to an individual due to radiation within the body from the quantity of radioactivity excreted. These interpretations should be based on a power function in cases where the retention of the assimilated radionuclide is known to be best represented by such a function. Formulae having the same parameters used in the appendix of the revised report[16] are derived in this report for use in making these interpretations.

RELATION OF QUANTITY EXCRETED TO THAT ASSIMILATED IN ACUTE EXPOSURES

In experimental observations where the power function has been found to describe retention of bone-seeking elements, the basic equation of retention (neglecting radioactive decay) is expressed as the power function

$$R(t) = At^{-n} \qquad t \geqq A^{1/n} \qquad 0 < n < 1 \qquad (1)$$

where $R(t)$ is the fraction of assimilated radioactive material retained t days postassimilation, A is a constant which is equal to the fraction of the assimilated dose retained when t is equal to 1 day, and $-n$ is a constant equal to the slope of the linear log–log transform of the retention function. It should be

noted that since this slope is negative, the power function increases without bound as t approaches zero. Therefore it has no meaning between the time of assimilation ($t = 0$) and some minimum value of $t(t = x)$ where the fraction retained equals unity. If $R(x) = 1$, then $Ax^{-n} = 1$ and $x = A^{1/n}$. From equation (1) the accompanying instantaneous rate of excretion is obtained by differentiating

$$\frac{dR(t)}{dt} = -Ant^{-(n+1)} \tag{2}$$

Substituting equation (1), the basic equation of retention, in equation (2) gives

$$\frac{dR(t)}{dt} = -\frac{nR(t)}{t} \tag{3}$$

The expression is negative since the fraction retained must always decrease. To the rate of loss by *excretion* may be added the rate of loss by *radioactive decay* to obtain the rate at which the assimilated material is *eliminated* from the body should the half-life be short. This gives

$$\frac{dR(t)}{dt} = -\frac{nR(t)}{t} - \lambda_r R(t) \tag{4}$$

where λ_r is the radiological decay constant. The fraction of the assimilated radioactive material retained, $R(t)$, when the radioactive decay is appreciable* can be obtained by integrating equation (4) giving

$$R(t) = At^{-n}\, e^{-\lambda_r(t-1)} \text{ (when } t > 1) \tag{5}$$

However, of interest to the bio-assayer is that fraction *excreted*. Thus the fractional *excretion* rate at time t, $Y(t)$, expressed as the fraction excreted per unit time, is the rate of *elimination* from the body at time, t less the rate of loss by *radioactive decay* or

$$Y(t) = -\frac{dR(t)}{dt} - \lambda_r R(t) \tag{6}$$

Substituting equation (4) and (5) in equation (6) gives

$$Y(t) = nAt^{-(n+1)}e^{-\lambda_r(t-1)} \tag{7}$$

The excretion rate at time t, $E(t)$, is equal to the product of the fractional excretion rate at time t and the quantity assimilated $q(o)$,
or

$$E(t) = q(o)\, Y(t) \tag{8}$$

* If the radiological half-life, T_r $(0.693/\lambda_r)$, is $\geqq 6.6t$, $0.9 < e^{-\lambda_r t} < 1.0$, and if $T_r \geqq 66t$, $0.99 < e^{-\lambda_r t} < 1.00$.

Substituting equation (7) in equation (8) and solving for $q(o)$ gives

$$q(o) = \frac{E(t)t^{n+1}}{nAe^{-\lambda_r(t-1)}} \tag{9}$$

It should be remembered that $dR(t)/dt$ in equation (6) is the instantaneous rate of elimination, and as such, should be integrated to determine the amount eliminated per day. However, this is unnecessary except at values of t close to the time of assimilation or when the unit of time considered is several weeks instead of 1 day, since the decrement in retention shortly becomes very small. When t is close to the time of assimilation or the unit of time over which the excreta is collected is long, the fraction of material which will be eliminated from the body during the period of time from t_1 to t_2 will be $R(t_1) - R(t_2)$. The fraction of the material which will be found in the urine and feces during this same period, Y, will be the fraction eliminated less that which has decayed due to radioactivity or

$$Y = R(t_1)e^{-\lambda_r(t_2-t_1)} - R(t_2) \tag{6'}$$

Substituting equation (5) in (6′) gives

$$Y = A(t_1^{-n} - t_2^{-n})e^{-\lambda_r(t_2-1)} \tag{7'}$$

Substituting equation (7′) in equation (8) and solving for $q(o)$ gives

$$q(o) = \frac{E}{A(t_1^{-n} - t_2^{-n})\, e^{-\lambda_r(t_2-1)}} \tag{9'}$$

where E is the quantity excreted during this period.

Should the quantity of material in the body at the time a sample was taken, $q(t)$, be desired, it can be calculated by substituting equation (5) and equation (9) in the simple relationship

$$q(t) = q(o)R(t) \tag{10}$$

giving

$$q(t) = \frac{E(t)t}{n} \tag{11}$$

Frequently an acute assimilation is discovered as the result of routine sampling, and later confirmed by a resample. The time of assimilation is usually not known and must be calculated before the quantity assimilated, $q(o)$, can be determined. Here equation (9) for the excretion rate of the first sample is

$$E(t_1) = \frac{q(o)nAe^{-\lambda_r(t_1-1)}}{t_1^{n+1}} \tag{12}$$

and for the second sample is

$$E(t_2) = \frac{q(o)nAe^{-\lambda_r(t_2-1)}}{(t_1 + \Delta t)^{n+1}} \tag{13}$$

where Δt is $t_2 - t_1$. Assuming that the exponential functions in these two equations are approximately equal, they can be solved simultaneously for t_1 giving

$$t = \frac{\Delta t}{\left(\frac{E(t_1)}{E(t_2)}\right)^{\frac{1}{n+1}} - 1} \tag{14}$$

Substituting this in equation (9) gives

$$q(o) = \frac{E(t_1)}{nAe^{-\lambda_r(t_1-1)}}\left[\frac{\Delta t}{\left(\frac{E(t_1)}{E(t_2)}\right)^{\frac{1}{n+1}} - 1}\right]^{n+1} \tag{15}$$

It should be noted here that the exponential term for radioactive decay, $e^{-\lambda r t}$, in the above equation is essentially unity for plutonium-239 and natural uranium. With radium-226 the value of the exponential term becomes less than unity when t becomes very large; however, it can never be less than 0.98 (the value when t is equal to an average occupational exposure of 50 years), and thus can also be considered unity. With strontium-90, $e^{-\lambda r t}$ becomes appreciably less than unity for large values of t. For values of t less than 4 years, however, it may be assumed to be unity without introducing an error greater than 10 per cent.

Table 1 gives the values which have been recommended for the constants used in the calculation of MPC values for soluble compounds and are applicable in the above equations.

Table 1. *Values recommended for power-function constants*

Nuclide	A	n	x*
$_{38}Sr^{90}$	0.65†	0.35†	0.29
	0.95†‡	0.254†‡	0.81
$_{88}Ra^{226}$	0.54†§	0.52‡§	0.31
$_{94}Pu^{239}$	0.99†	0.01†	0.37
Natural uranium	0.72†	0.80†	0.66
	0.60**	0.50**	0.36

* $x = A^{1/n}$

† ICRP Sub-Committee on Permissible Dose for Internal Radiation.

‡ MARINELLI[21]

§ NORRIS[7]

** BERNARD[15]

In the preceding equation the term $E(t)$ refers to the total amount excreted per unit time. This includes material eliminated via the urine, $E_u(t)$, and via the feces, $E_f(t)$. Consequently $E(t)$ must be calculated from either one or both of the above terms before it can be used in these calculations.

Generally, data on the rate of urinary elimination are more available and the total amount excreted is calculated from it. (This may be done even when data on fecal elimination are available owing to the difficulty in establishing the period of time represented by a single defecation and the fluctuations in the quantity of radioactivity in successive samples.) The total excretion rate for plutonium compounds readily accessible to circulating body fluids at time t can be calculated from the urinary excretion rate alone using the formula

$$E(t) = 4E_u(t)t^{-0.2}, \quad \text{when} \quad t < 1024 \tag{16}$$

This can be calculated from data provided by LANGHAM[12]. BERNARD[15] found that the excretion of uranium takes place largely in the urine. The fraction of the daily excretion from humans injected with soluble compounds which appeared in the feces ranged from 1.1×10^{-3} to 6.6 per cent but seldom exceeded 1 per cent. Thus all of the uranium accessible to the circulating body fluids may be considered to be excreted in the urine. In contrast to uranium, the excretion of radium takes place largely in the feces. The average value for the radium excreted in the urine probably lies between 1.5 and 5 per cent.[17] There also appears to be no trend of this ratio as a function of time after radium assimilation as in the case of plutonium. The ratio for a single day's sampling may vary several-fold from individual to individual and in the same individual. Therefore, inferences about the daily excretion rate based on a single 24-hr urine sample will be, at best, approximations.

Very little systematic study of strontium metabolism in man is available. Clinical investigation by HARRISON[18] using nonradioactive strontium and by LASZLO, SPENCER *et al.*[19,20] using strontium-85 have resulted in some data which can be used to relate urinary to fecal excretion. For the first 5 days following intravenous injection of soluble strontium, the main avenue of excretion is via the urine. However, as the urinary excretion rapidly drops, the fecal excretion increases. For times longer than 5 days, roughly 60 per cent of the daily excretion is urinary. About 80 per cent of the orally ingested soluble strontium remains unabsorbed and is eliminated in the feces over a 6-day period. After this time roughly 80 per cent of the daily excretion is urinary.

Table 2. Change in ratio of two plutonium-retention equations with time

t (days)	$Y(t)/Y_{u+f}$
1	1.25
10	1.07
100	0.91
1000	0.77

It might be explained that the power function formulae used for plutonium excretion rates in the past[11–13] cannot be used to relate retention to excretion, since their negative exponents are less than unity and the integral of such a function is divergent and has no value. However, using the above constants, the negative differential of the retention equation for plutonium,

$$R(t) = 0.99t^{-0.01} \tag{17}$$

is

$$-\frac{dR}{dt} = 0.0099t^{-1.01} = Y(t) \tag{18}$$

This is not too different from the equation previously used by LANGHAM[11]

$$Y_{u+f} = 0.0079t^{-0.94} \tag{19}$$

These two equations may be further compared by taking their ratio

$$\frac{Y(t)}{Y_{u+f}} = \frac{0.0099t^{-1.01}}{0.0079t^{-0.94}} = 1.25t^{-0.07} \tag{20}$$

The numerical values of this ratio with increasing time are given in Table 2.

CALCULATION OF INTEGRAL ABSORBED DOSE IN AN ACUTE ASSIMILATION

Having estimated the quantity assimilated into the body as the result of a single acute exposure, $q(o)$, it is usually desirable to assess the relative dose the body will receive from the ionizing radiation. It should be noted that the values given in the literature[22,23] for q* are calculated on the premise that the accumulation of the burden in the critical body organ is due to the exclusive use of air or water contaminated with a fixed concentration of the nuclide for a time that is much longer than the effective half-life of the nuclide in the critical body organ, but not greater than 50 or 70 years. These values are not comparable in the case of a single acute assimilation. In assessing these cases, the average dose rate in rems per unit of time over a specified interval of time, viz. rems per week for the first week, must be calculated from $q(o)$.

The RBE dose in rems can be defined as the quotient of the integral absorbed RBE dose divided by the mass of either the critical body organ or the whole body, depending on the parameters used. The unit of integral absorbed RBE dose is the gram-rem which is the product of the integral absorbed dose in gram-rads[24] and an agreed conventional value of the RBE with respect to a particular form of radiation effect. Since radiation dosage accumulates as the product of concentration of the radioelement and time, the integrated dosage* over t days, $D(t)$, is

$$D(t) = q(o)Kf(R) \tag{21}$$

where $q(o)$ is the quantity assimilated in disintegrations per minute. The quantity $f(R)$ is in every case the appropriate retention function integrated with respect to t over the period in question. K is the radiation dosage rate per day per disintegration per minute.

$$K = \frac{(1440)(1.6 \times 10^{-6})\,\Sigma E(\text{RBE})N}{100}$$
$$= 2.3 \times 10^{-5}\,\Sigma E(\text{RBE})N \tag{22}$$

where

1440 = min/day;
1.6×10^{-6} = ergs/MeV;
ΣE = effective energy per disintegration (MeV);
RBE = relative biological effectiveness (rems/rad);
N = relative damage factor;
100 = ergs/gram-rad.

* The maximum permissible body burden of a radioactive nuclide in a standard man.

* The integrated dosage should not be confused with the integral absorbed dose which is related to the dose in rems as follows:

$$\text{dose (rems)} = \frac{\text{integral dose}}{\text{weight of tissue}}.$$

Substituting equation (5) for the quantity $f(R)$ in equation (21) gives

$$D(t) = q(o)KA\int_x^t \tau^{-n}e^{-\lambda_r(\tau-1)}\,d\tau \qquad (23)$$

where x is that fraction of a day when the fraction retained is one and is numerically equal to $A^{1/n}$. This is the Laplace transform of the function τ^{-n} and can be solved by converting it to the form

$$D(t) = \frac{q(o)KAe^{\lambda_r}}{\lambda_r{}^{(1-n)}} \times [\mathrm{I}(u,p) - \mathrm{I}(u',p)]\,\Gamma(1-n) \qquad (24)$$

Where the arguments to the function $\mathrm{I}(u,p)$ are $p = -n$, $u = \lambda_r t/\sqrt{(1-n)}$, and $u' = \lambda_r x/\sqrt{(1-n)}$. The numerical values for the function $\mathrm{I}(u,p)$ are given in Tables 2 and 3 of *Tables of the Incomplete* Γ*-Function.*[25] The numerical values for the function $\Gamma(1-n)$ can be computed from any table of the Γ-function using the relationship

$$\Gamma(1-n) = \frac{\Gamma(2-n)}{1-n} \qquad (25)$$

Should tables of the incomplete Γ-function not be available, $D(t)$ can be calculated from the infinite series

$$D(t) = \frac{q(o)KAe^{\lambda_r}}{\lambda_r{}^{1-n}} \times \sum_{j=1}^{\infty} \frac{(\lambda_r t)^{j-n}e^{-\lambda_r t} - (\lambda_r x)^{j-n}e^{-\lambda_r x}}{j-n} \qquad (26)$$

Where the radiological half-life is very long and λ_r is insignificant

$$D(t) = \frac{q(o)KA}{1-n}\,(t^{1-n} - x^{1-n}) \qquad (26')$$

and can be obtained from either equation (23) or equation (26).

However, should this equation be used to calculate the integral absorbed dose from strontium-90 over the average occupational exposure of 50 years, D (50 years) would be 175 per cent of the true value.

RELATION OF QUANTITY EXCRETED TO RATE OF ASSIMILATION IN MULTIPLE OR CONTINUOUS EXPOSURES

In the case of multiple assimilations of dissimilar quantities of a radioactive nuclide or of similar quantities assimilated at long or dissimilar intervals, estimates of retained amounts must be derived by summation of individual items over the appropriate time intervals. However, this may be impossible if, as is often the case, information on the time of the assimilations or the quantity previously assimilated is lacking. Here it is necessary to assume a chronic invariant assimilation. This is a hypothetical case in which an individual assimilates a radioactive nuclide continuously at a fixed rate, q, over an interval of time T. There are instances, however, in which such hypothetical cases are approached, in that radioactive nuclides are made available to the body from its own metabolic pool (e.g. the lungs), at very slow rates. However, when repeated assimilations of similar magnitude occur at reasonably short and constant intervals, the resulting picture can be closely approximated by the chronic invariant case.

If the excretion rate resulting from each increment assimilated is described by equation (9),

$$E(t) = q(o)nAt^{-(n+1)}e^{-\lambda_r(t-1)}$$

the total excretion on day t resulting from a continuous assimilation, $E_c(t)$, can be considered as the sum of the excretion rates from each of the incremental steps or

$$E_c(t) = qnA\int_x^T (t-\tau+x)^{-(n+1)} \times e^{-\lambda_r(t-\tau+x-1)}\,d\tau \qquad (27)$$

where the time following assimilation in equation (9) is given by the difference between the time of excretion t and τ, the time of each incremental assimilation. The constant x, which is the time when the fraction retained is unity, is added here since, according to the previous model, no excretion occurs during times less than x. Thus the values of the time following assimilation will never be less than x, and will approach $t-\tau$ asymptotically as t increases. This equation cannot be solved using the present tables of incomplete Γ-functions, but can be expressed as the infinite series

$$E_c(t) = -qnAe^{-\lambda_r(t-1)} \times \sum_{j=0}^{\infty} \frac{\lambda_r{}^j(t-T+x)^{j-n} - t^{j-n}}{(1-n)(2-n)\cdots(j-n)} \qquad (28)$$

Where the radiological half-life is very long and λ_r is insignificant

$$E_e(t) = qA[(t - T + x)^{-n} - t^{-n}] \quad (29)$$

can be obtained from either equation (27) or equation (28).

CALCULATION OF QUANTITY ASSIMILATED IN MULTIPLE OR CONTINUOUS EXPOSURES

Generally, with chronic assimilations the values for q, t and T are not known. However, if two measurements are made of the quantity of material excreted at time t_1 and t_2, where $t_2 = t_1 + \Delta t$, the total quantity assimilated can be calculated by solving equation (29) for t. In order to do this the term $(t - T + x)^{-n}$ must be expanded. This can be done by considering it as a binomial, $[t - (T - x)]^{-n}$, and expanding it using the binomial series. Since the exponent is not a positive integer this will result in an infinite series which is approximated here by using only the first two terms. Thus,

$$(t - T + x)^{-n} \cong t^{-n} + n(T - x)t^{-(n+1)} \quad (30)$$

From the above relationship it can be seen that as T approaches t the approximation becomes poorer. Therefore a minimum value for the ratio of $t - T + x$ to t, below which this approximation will not be valid, must be established for each element. Table 3 gives the values for the ratio of $t - T + x$ to t above which the approximation of the binomial will be better than 90 per cent.

These ratios were calculated by relating the approximate formula to the binomial as a per cent or

$$\% = \frac{[t^{-n} + n(T - x)t^{-(n+1)}]\ 100}{(t - T + x)^{-n}} \quad (31)$$

Setting $T - x = 1$, the values for t which would give 90 per cent were found by trial and error. These values of t were then substituted in

$$\frac{t - 1}{t}$$

to give the above ratios. It can be seen from Table 3 that the approximation may be used to describe the excretion of plutonium at any time t; however, some discretion must be used with the other elements. Substituting this approximation [equation (30)] in equation (29) gives

$$E_e(t) = qAn(T - x)t^{-(n+1)} \quad (32)$$

Substituting t_1 and t_2 in the preceding equation and solving the two equations simultaneously for $q(T - x)$ similar to the derivation of equation (15) gives

$$q(T - x) = \frac{E_e(t_1)}{nA}\left[\frac{\Delta t}{\left(\frac{E_e(t_1)}{E_e(t_2)}\right)^{\frac{1}{n+1}} - 1}\right]^{n+1} \quad (33)$$

This formula represents the quantity of material assimilated due to a chronic invariant exposure since it is the product of the quantity assimilated daily and the period of the exposure in days. It is particularly significant here that, using this formula, the total quantity assimilated can be calculated when only the quantity excreted on two different days and the time interval between the excretions are known.

Table 3. Values for the ratio $(t - T + x)$ to t above which the approximation of the binomial is better than 90 per cent

Nuclide	n	$\frac{t - T + x}{t}$
$_{94}Pu^{239}$	0.01	0.0001
$_{38}Sr^{90}$	0.35	0.45
Natural uranium	0.50	0.55
$_{88}Ra^{226}$	0.52	0.55

CALCULATION OF INTEGRAL ABSORBED DOSE IN MULTIPLE OR CONTINUOUS EXPOSURES

For the sake of simplifying the derivations, only the hypothetical chronic invariant assimilation discussed earlier will be considered. In this case, if the dose resulting from each increment assimilated is described by equation (26′)

$$D(t) = \frac{q(o)KA}{1 - n}\,(t^{1-n} - x^{1-n})$$

where $K = 2.3 \times 10^{-5}\ \Sigma E(\mathrm{RBE})N$, the total dose can be considered as the sum of the doses from each of the incremental steps. In setting up this equation, the time since assimilation in equation (26′) must be taken as the difference between the time of observation t and τ, the time of assimilation, or $(t - \tau + x)$. The reason for the constant x has been discussed earlier. Integrating equation (26′) over the period of exposure, T,

$$D_c(t) = \frac{qKA}{1-n} \times \int_x^T [(t - \tau + x)^{1-n} - x^{1-n}]\, d\tau \quad (34)$$

gives

$$D_c(t) = \frac{qKA}{n^2 - 3n + 2}[t^{2-n} - (t - T + x)^{2-n}] - \frac{qKAx^{1-n}}{1-n}(T - x) \quad (35)$$

Where the radionuclide is Pu^{239} and n is a small number, equation (35) can be approximated by

$$D_c(t) \cong q(T - x)KA\left(\frac{2t - T - x}{2}\right) \quad (36)$$

Although a value for $q(T - x)$ may be obtained from equation (33), this formula, as well as equation (35), will be of little value unless some information on t and T is available. Therefore, a better appraisal of the magnitude of the damage resulting from a multiple or continuous assimilation of these nuclides, particularly if t and T are large, may be made by comparing the quantity assimilated, calculated using equation (33), with the values of the maximum permissible total body burden for a 70-kg man found in the literature.[22,23]

SUMMARY

If a power-function formula is used to describe the retention of radionuclides in the body, the relationship between the quantity assimilated in an acute exposure $q(o)$ and that excreted at time t days postassimilation, $E(t)$, is given by

$$q(o) = \frac{E(t)t^{n+1}}{nAe^{-\lambda_r(t-1)}}$$

where A is a constant which is equal to the fraction of the assimilated material retained when t is equal to 1 day, n is a constant equal to the negative slope of the linear log–log transform of the retention function, and λ_r is the radiological decay constant. When the values of t are close to the time of assimilation or when the unit of time, $t_1 - t_2$, over which the excreta is collected is long, the relationship will be better represented by

$$q(o) = \frac{E}{A(t_1^{-n} - t_2^{-n})e^{-\lambda_r(t_2-1)}}$$

Whereas in the past, power-function equations have been given for urinary or fecal excretion alone, the quantity expressed here by $E(t)$ or E represents the total excretion. Therefore, if only urinary excretion data are available, the value of $E(t)$ or E must be extrapolated based on experimental data.

If multiple assimilations are of similar magnitude and occur at reasonably short and constant intervals, and if the radiological decay constant is approximately zero, the relationship between the quantity assimilated per unit time, q, over a period of T days and $E_c(t)$, the quantity excreted at time t days after the commencement of the exposure can be given by

$$E_c(t) = qA[(t - T + x)^{-n} - t^{-n}]$$

where x is that fraction of a day when the fraction retained is unity.

Frequently, the fact that a radionuclide has been assimilated is discovered as the result of routine sampling and later confirmed by a resample. Here the quantity assimilated as the result of an acute exposure can be calculated using the relationship

$$q(o) = \frac{E(t)}{nAe^{-\lambda_r(t_1-1)}}\left[\frac{\Delta t}{\left(\frac{E(t_1)}{E(t_2)}\right)^{1/(n+1)} - 1}\right]^{n+1}$$

where $E(t_1)$ and $E(t_2)$ are the quantities excreted on the two different occasions Δt days apart. Within the limits shown in Table 3, the relationship between the quantity of a radionuclide assimilated over a period of $T - x$ days,

$q(T-x)$, and that excreted on two different occasions can be given by

$$q(T-x)=\frac{E_c(t_1)}{nA}\left[\frac{\Delta t}{\left(\frac{E_c(t_1)}{E_c(t_2)}\right)^{1/(n+1)}-1}\right]^{n+1}$$

The calculation of the integral absorbed dose due to a chronic invariant assimilation requires information which in most cases is not available and must be approximated. Therefore, the magnitude of the damage resulting from such an exposure may be better appraised by comparing the quantity assimilated with the values of the maximum permissible total body burden for a 70-kg man found in the literature.[22,23] The integral absorbed dose in gram-rems over a period of time t, which is due to an acute exposure, may be calculated from the equation

$$D(t)=\frac{q(o)KA}{1-n}\,(t^{1-n}-x^{1-n})$$

where $K=2.3\times10^{-5}\,\Sigma E(\mathrm{RBE})N$. When the radiological decay constant is appreciable the equation

$$D(t)=\frac{q(o)KAe^{\lambda_r}}{\lambda_r^{(1-n)}}\left[\mathrm{I}(u,p)-\mathrm{I}(u',p)\right]\Gamma(1-n)$$

must be used. Here the arguments to the function $\mathrm{I}(u,p)$ are $p=-n$, $u=\lambda_r t/\sqrt{(1-n)}$, and $u'=\lambda_r x/\sqrt{(1-n)}$. The numerical values for the function $\mathrm{I}(u,p)$ are given in Tables 2 and 3 of *Tables of the Incomplete* Γ*-Function*,[25] and the values for the function $\Gamma(1-n)$ can be computed from any table of Γ-functions using the relationship

$$\Gamma(1-n)=\frac{\Gamma(2-n)}{1-n}$$

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NOTES

Low-Background Shields*

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WHEN counting γ-rays at very low levels, it is necessary to reduce the background by shielding out the γ-rays from the environment. It is obvious that the lower the amount of activity to be counted, the lower the background must be. Today in many applications, it is desired to measure as small activities as possible, e.g. counting of the natural activity of the human body. It is of interest to consider the type of shield to be used.

In the laboratory where relatively small shields are used, lead is perhaps the most common material. In other applications where large shields are needed, iron[1] and water[2,3] have been used. All of these materials have certain advantages and disadvantages such as inherent amounts of radioactivity (mostly K^{40} and radium), cost and ease of construction. Let us first consider the criterion to be used to decide if the amount of γ-ray activity present in the shield is sufficiently low.

We can idealize our shield as a sphere of inner radius a and outer radius b made of material with an absorption coefficient μ' cm^2/g, emitting q γ-rays/sec per g, and with a density ρ. Then with the counter at the center of the sphere, the number of γ-rays/cm^2 per sec from a spherical shell at distance r and thickness dr is given by

$$dn = \frac{4\pi r^2 q\rho e^{-\mu'\rho(r-a)}}{4\pi r^2}\,dr \tag{1}$$

and the total number of γ-rays from the entire wall is

$$n = q\rho \int_a^b e^{-\mu'\rho(r-a)}\,dr \tag{2}$$

$$n = \frac{q}{\mu'}\left[1 - e^{-\mu'\rho(b-a)}\right] \tag{3}$$

We shall limit our discussion to shields sufficiently thick that the second term of the bracket is essentially zero and the contribution of the γ-rays from outside the shield is also negligible. In this case

$$n \approx \frac{q}{\mu'} \tag{4}$$

q/μ' can be considered a figure of merit for a shielding material and should be made as small as possible. However, in comparing shielding materials of high and low atomic numbers (Z), a different value of μ' must be chosen. In high Z materials, the photoelectric absorption cross-section is very high for low-energy γ-rays and γ-rays degraded in energy by Compton scattering will be quickly absorbed. In low Z materials this is not the case and many of the degraded γ-rays will penetrate to the counter. This is shown experimentally by the fact that a thin Pb liner in a low Z shield will always reduce the background.[4] A value for μ' should then be chosen ranging from nearly the total attenuation cross-section for high Z materials such as lead, down to little more than just the Compton absorption cross-section for low Z materials such as water. Thus for aluminum, a value of $\mu' = 0.03$ cm^2/g might be chosen at both 1 MeV and 0.2 MeV while for lead $\mu' = 0.07$ cm^2/g and $\mu' = 0.95$ cm^2/g would be appropriate at these energies. If the γ-rays present had an energy of 1 MeV the lead could have 2.3 times as much activity per gram present as Al and still be as good a shield while at 0.2 MeV, over 30 times as much activity could be present in the lead and be an equally good shield.

Radium (average γ-ray energy $\sim$0.8 MeV) and K^{40} (γ-ray energy 1.46 MeV) are the usual contaminants present. Over this range of energies, we can use $\mu' \approx 0.03$ cm^2/g for all low and medium Z materials and $\mu' \approx 0.07$ cm^2/g for Pb. For typical materials in the surroundings, radium contents of about 10^{-12} g Ra/g are found. Al contains about 3×10^{-13} g Ra/g,[1] steel $< 10^{-14}$ g Ra/g,[5] and some tap water $< 10^{-16}$ g Ra/g.[5] It can be seen that widely differing figures of merit can be obtained for different shield materials. For very thick shields, however, RASMUSSEN[5] found about the same background for a NaI scintillation counter for materials of widely differing radium content. This indicates that most of the residual background in any good shielding material (e.g. iron, water or lead) is due to γ-rays from the scintillation crystal, housing and photomultiplier tube. Unless this contribution is reduced, it is adequate to find any material with total activity of $\sim 10^{-14}$ c/g or less. Since several materials satisfy this requirement, the factor determining one's final choice will be cost, ease of construction, etc.

Of the common materials, lead is probably the most expensive. Iron and water are about equally

* This paper is based on work performed under Contract W-31-109-eng-52 for the Atomic Energy Commission.

expensive when the cost of tanks to contain the water is included. Iron is probably the easiest to use to construct large shields; and for small shields, lead bricks are very convenient.

It has been felt that there might exist some other raw materials which would be adequate shields and would be cheaper than iron or lead.[5] A search was made in this laboratory for some commercially produced rock or mineral which would be suitable. Many samples of the ultrabasic rocks produced in Washington and Oregon were tested.* Samples of ~2 lb were placed near the face of a 5 in. × 2 in. NaI(Tl) crystal inside a lead shield with 4-in. thick walls and an integral γ-ray count was made. Those samples giving no increase in background, were obtained in 100-lb samples and packed all around the Na(Tl) crystal for a more sensitive test. Three of these 100-lb samples gave good results; one of peridotite and two of talc. All of these rocks came from Skagit County in Western Washington. The talc was chosen for further testing because it is a soft rock easily crushed. Thirty tons of this were obtained in polyethylene-lined paper bags containing 100 lb each. From this, a cell was constructed with inner dimensions 4 ft × 4 ft × 4 ft and 2-ft thick walls. A sketch of the cell is shown in Fig. 1. Construction was made by simply piling the bags up to form walls and laying iron sheets across the top to support the roof. A steel jack post helps to support the roof and makes a convenient support for the detector. Two of the walls were against the walls of the room and required no other support. The third wall was made double thickness for stability. The fourth wall was a rolling door, again 2 ft thick. This door was made by stacking the bags of rock on a ¼-in. iron plate. Channel iron tracks were placed on the floor to provide a hard surface. Three 1-in. steel rollers were placed between the plate and the tracks. The rollers were pinned at each end to a piece of light angle iron to keep them parallel while constructing the door. Although this door rolls quite easily despite its weight (~2¼ tons), a motor drive was added for the use of a girl operator. The motor pulls the door closed up a slight incline and allows it to coast open.

Background spectra were taken in this cell with a 5 in. × 2 in. NaI(Tl) crystal and compared with the background in a small shield with 4 in. Pb walls. The results are shown in Fig. 2. It can be seen that the background in the talc cell ranged from about 200 per cent at low energies to 160 per cent at high energies of that obtained in the lead cell.

* The writer wishes to express his appreciation to Mr. R. E. Brown, Hanford Laboratories, for his help in procuring these samples.

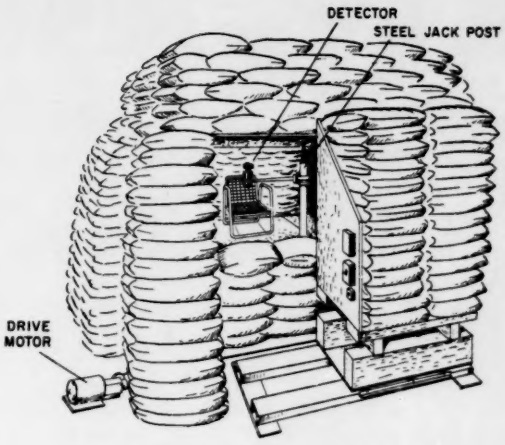

Fig. 1. A low-background shield made of crushed talc.

A flame photometer analysis of the talc yielded a potassium content of 25 p.p.m. or about 2×10^{-15} c/g of γ-ray activity. A radium analysis made by de-emanation of powdered talc and a subsequent radon count gave from 4 to 8×10^{-15} c radium/g of talc.* The uncertainty is due to the unknown efficiency of de-emanation. The background shown in Fig. 2 is consistent with a radium spectrum. By placing another photomultiplier and preamplifier in front of the crystal, it was possible to estimate that about 18 per cent of the background came from the measuring equipment itself. This background was not

* The writer wishes to thank the MIT Radioactivity Center for making this measurement.

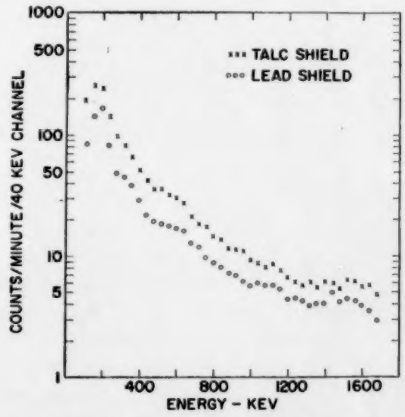

Fig. 2. Background spectra obtained in the talc shield and in a lead shield.

as low as had been hoped, but is adequate for most purposes. At present the cell is being used for body γ-ray measurements.

In addition to the higher background, the talc cell is also much bulkier than a lead-walled cell with the same internal dimensions. The 2-ft thick walls were required to give the same amount of absorption to external γ-rays as 4 in. of lead. The cost of this cell, however, was only about 20 per cent of that of the lead cell, or about $1100, including all material and construction and shipping costs.

This type of cell should be of interest in applications where the higher background is tolerable and economy is desirable. It has been suggested that enough of the talc be stockpiled at various plants using radioactive materials, so that a low-background human counting facility could be quickly set up in case of an accident. There are undoubtedly other rocks in other parts of the country which would also be suitable. RASMUSSEN[5,6] has found considerable promise in dunite, which is common on the east coast.

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On the Present Possibilities of Washing Radioactive Substances off the Skin of Living Animals

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IT WAS the purpose of our experimental work to prepare the most efficacious combination of substances suitable for the decontamination of living skin contaminated with long-life fission products of U^{235}. On extensive material (shaved skin of living rabbits, pigs, men) we have proved experimentally that the most suitable mixture for decontamination consists of a detergent and sodium salts of EDTA acid, or of a detergent and sodium salts of polyphosphoric acids. When this had been determined we posed another question, namely whether the decontamination agent containing EDTA and the decontamination agent containing the polyphosphoric acids are equivalent as to their effect. From the two substances the following decontamination agents were combined:

(I) Sodium salts of polyphosphoric acids, Graham's salt (pulverized), 10.0 g; sodium salt of laurylsulfonic acid, 2.5 g; crystalline sodium carbonate (pulverized) 85.0 g. Five grams of this mixture was added to 100 ml of water.

(II) Disodium salt of EDTA acid (pulverized), 5.0 g; sodium salt of laurylsulfonic acid, 5.0 g; electrophoretically purified starch, 5.0 g; crystalline sodium carbonate (pulverized), 85.0 g. Ten grams of this mixture was added to 100 ml of water.

Shaved, otherwise intact, dried skin of living laboratory animals was deprived of fat on the surface by alcohol-ether and contaminated with two to three drops of the contaminating solution over an area of 4 cm^2. This solution contained, on the one hand carriers, on the other hand soluble salts of the following elements: Cs^{134}, $Sr^{90} + Y^{90}$, $Ce^{144} + Pr^{144}$, $Zr^{95} + Nb^{95}$, $Ba^{140} + La^{140}$, Co^{60}, I^{131}. The elements were combined in such mutual relations as occur in one-year-old fission products of U^{235}. After contamination, the skin of the animals was dried by means of an infra radiator and its radioactivity was measured with theoretical effectiveness of 5 per cent by application of a beta GM tube with a thin window (4.0–5.7 mg/cm^2, 28 mm dia.). The contaminating solution was diluted with water so that the GM tube registered, at a distance of 3 cm from the skin surface, the lowest value of 10,000 impulses/min and the highest value of 15,000 impulses/min. The decontamination of the skin was performed by rubbing of the skin for 1 min with a soft tampon abundantly soaked with the decontaminating solution. Then the skin was washed with a stream of running water. This process of decontamination was repeated three times successively. The skin was then dried with a tampon and by means of an infra radiator, after which a radiometric examination was performed again.

From the originally applied activity only 3.0 $\pm$1.2 per cent remained on the skin if decontamination was performed within 1 hr following contamination. If the skin was decontaminated as late as 4 hr after the contamination, 4.2 $\pm$0.8 per cent of the original activity remained on it. If the skin was decontaminated 8 hr after the contamination, as much as 6.3 $\pm$1.8 per cent of the originally applied activity remained on it.

In 728 experiments on shaved skin of living pigs, rabbits and men, we proved that the decontamination agents (I) and (II) are equivalent. We prefer, however, the agent (I) because it is cheaper, simpler, and its manufacture is easier. Apart from decontamination of living skin it can be also used for washing of contaminated textiles.

The efficacy of the solutions (I) and (II) combined by us, was compared also, for the purpose of control, with the decontamination effect of the single components of these solutions alone. Decontamination performed with pure water and washing left up to 20 per cent of the originally applied activity on the skin, decontamination with mere soda solution left 12 per cent, decontamination with the solution of the detergent left only 10 per cent, and decontamination with the complex forming substance left 8 per cent. We have proved that the *combination of a detergent, a complex forming substance and a chemical, adjusting suitably the* pH *of the solution, is the most efficacious for the decontamination of living skin.*

For the purpose of control we further compared experimentally the effectiveness of the decontamination agents (I) and (II) with the effectiveness of other decontamination agents containing: tartaric acid, citric acid, triethanolamine, various textile soaps, kaolin, TiO_2, diluted mineral acids. Decontamination agents consisting of these substances were always 6–15 per cent less effective than the decontamination agents (I) and (II). Besides, the first two substances mentioned bring with them the danger of increased penetration of the radioactivity, left on the skin, deeper into the organism. Triethanolamine was toxic, and diluted mineral acids injured the skin. We have therefore rejected all these agents.

We have further proved that the degree of decontamination by means of the solutions (I) and (II) becomes higher if a thin layer, invisible to the eye, of cold-cream is applied to the skin before the contamination. Under these conditions only 1.8 $\pm$0.5 per cent remained on the skin after decontamination.

The radioactivity which remained on the skin after decontamination with the agents (I) and (II) could always be subsequently removed by a short application of a keratolytic (BaS) followed by a fresh washing of the skin with the decontamination solution (I) or (II) and afterwards with water. We cannot, however, recommend this procedure because it damages the anatomical integrity of the skin. We only mention it to prove that the radioactivity which had not been washed away remained in the highest superficial horny layer of the skin.

Histological examinations of the decontaminated skin did not show any morphological changes. Healing of a deep skin wound, which the decontamination solutions (I) and (II) penetrated, took the normal course, the same as in a control wound. No allergic reaction of the skin, either in animals or in men, could be observed on the decontamination agents (I) and (II).

The decontamination solutions (I) and (II) did not enable a further accelerated penetration of radioactivity through intact skin. On correlation with animals whose skin had been washed only with water we did not find any increase in the radioactivity of the blood or the organs after decontamination of the skin with the agents (I) and (II).

In this work we have proved experimentally that the decontamination agents (I) and (II) are reciprocally equivalent and that they are, at present, most suitable for the decontamination of living skin contaminated with fission products of U^{235}. The decontamination agents (I) and (II) do not damage living skin either functionally or anatomically, and neither do they carry radioactivity deeper into the skin.

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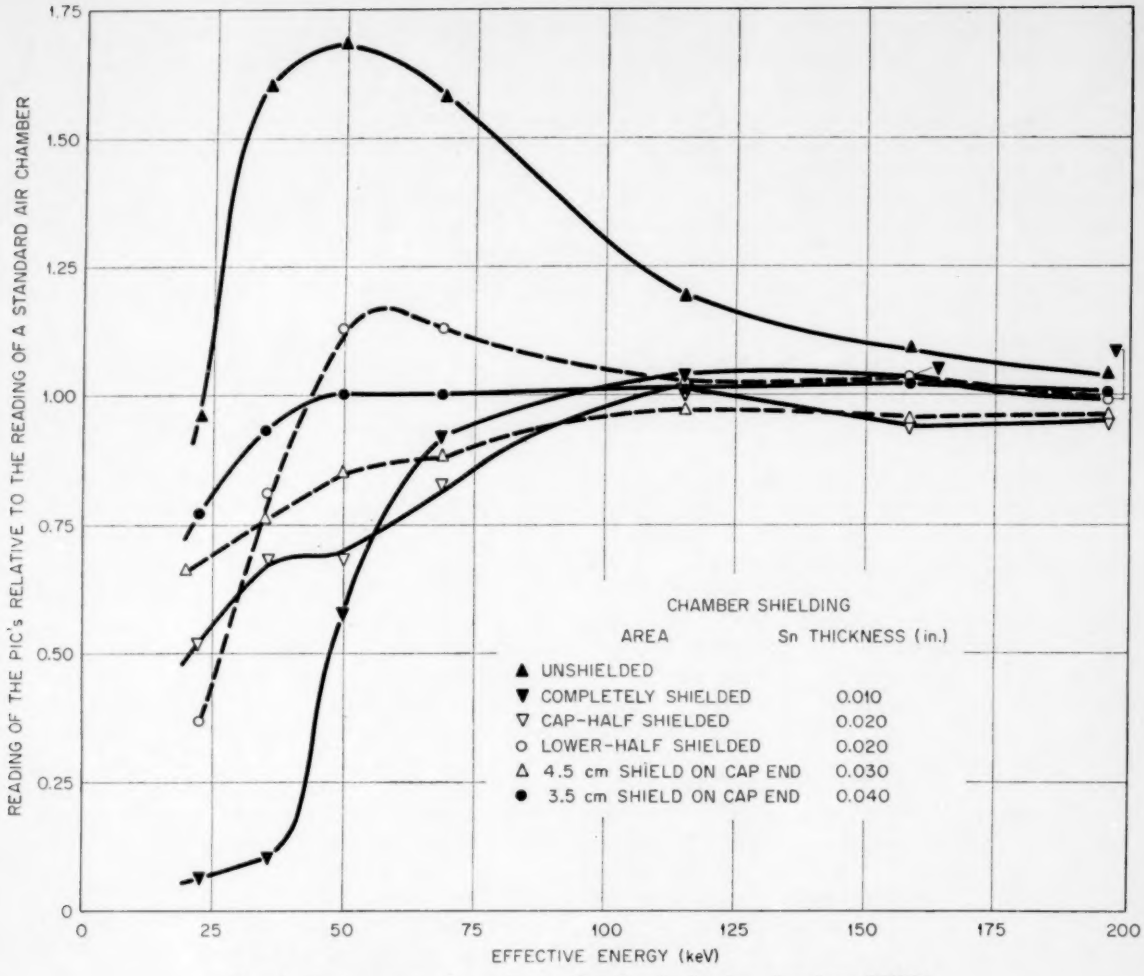

FIG. 1. X-ray energy dependence of pocket ionization chambers (PIC's).

A Simple Method of Minimizing the Energy Dependence of Pocket Ionization Chambers

(*Received* 17 *November* 1959)

ACCURATE measurements with pocket ionization chambers have been limited to known spectra because of their energy dependence.[1] However, the reproducibility and convenience of use of these instruments make them desirable for use in many applications. A simple modification of these instruments which would correct the response over a wide range of energies would increase their utility. A series of measurements were made with Victoreen Model 362 chambers with tin sleeves of several thicknesses and lengths covering all or part of the instrument. A modified Westinghouse Quadrocondex 250 KVP X-ray machine, fitted with appropriate filters was used for the low-energy exposures. A Victoreen condenser r-meter calibrated by comparison with a standard air chamber was used for X-ray measurements. The effective energy* was well known.[2]

Response of the chambers at higher energies was checked with Cs^{137} and Co^{60} sources. Fig. 1 summarizes the results of the X-ray measurements. A

* The effective energy of a heterochromatic X-ray beam is the energy of a monochromatic beam which has the same absorption coefficient as the given beam in an incremental thickness of standard filter material.

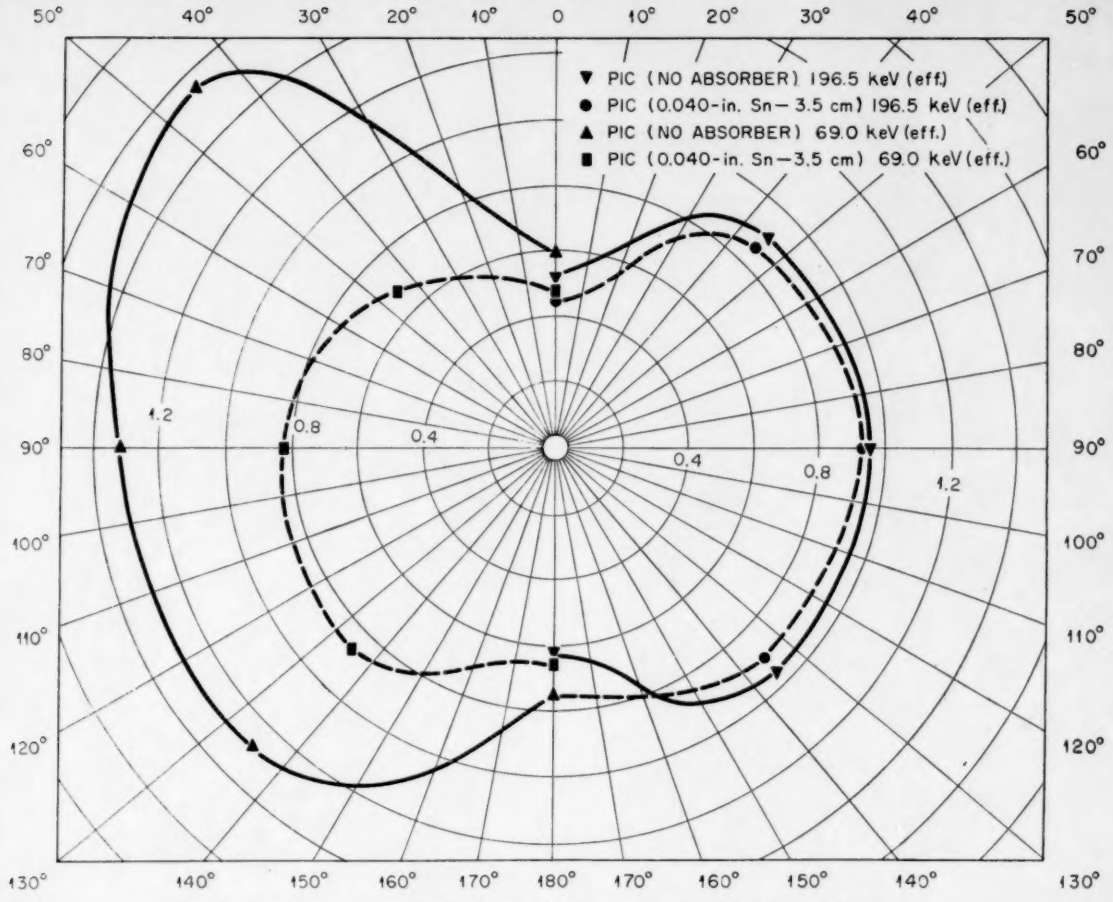

FIG. 2. Angular response of pocket ionization chambers (PIC's) to X-rays.

tin shield of 0.040 in. thickness, covering 3.5 cm of the charging or cap end of the chamber, yielded the most satisfactory response. Deviations of the response relative to the standard-air-chamber response were less than 10 per cent for energies above 35 keV. It was noted that the response is less sensitive to the thickness of the shield than to the portion of the chamber covered. Therefore, other thicknesses and/or materials could be expected to give a similar response correction.

The response of the chamber as a function of orientation of the central axis, relative to the direction of the incident radiation, was measured with and without the tin filters. Fig. 2 illustrates the results for bare and for shielded chambers for two effective energies of X-rays, 196.5 and 69.0 keV. In Fig. 2, 0° is that orientation in which the X-ray beam entered the chamber through the charging or cap end.

The use of a 0.040-in. tin filter, or sleeve, over 3.5-cm length of the charging end of a pocket ionization chamber permits more extensive use of this versatile instrument. The tin is easily wrapped around or slipped over the end; pressure by the fingers will usually be sufficient to indent the tin to hold it on, or it can be held in place with tape.

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*Oak Ridge National Laboratory** J. A. AUXIER
Oak Ridge, Tennessee J. S. CHEKA

References

1. F. H. DAY, *Ra-Det* **3**(4), 1 (1950).
2. J. C. VILLFORTH, R. D. BIRKHOFF and H. H. HUBBELL, Jr., *Comparison of Theoretical and Experimental Filtered X-Ray Spectra.* ORNL-2529 (1958).

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Energy Commission.

Health Physics Pergamon Press 1960. Vol. 2, p. 310. Printed in Northern Ireland

LETTER TO THE EDITOR

Suggested Insignia for the Health Physics Society

(*Received* 15 *September* 1959)

THE figures below are suggested insignia for the Health Physics Society. The globe suggests universality of the Society, the pinwheel suggests radiation and the helium nucleus indicates the atomic age. These insignia cannot be attributed to any one individual as numerous people have contributed the ideas.

J. C. HART

Health Physics Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee

Health Physics Pergamon Press 1960. Vol. 2, pp. 311–315. Printed in Northern Ireland

NEWS

Health Physics in the News

THE following items concerning Health Physics have appeared in newspapers, public press or technical journals during the last quarter.

Radioisotope training program

The AEC has announced a new program which will give students of small undergraduate colleges opportunity for specialized training in the techniques of using radioisotopes. The program will utilize a mobile laboratory for presentation of a two-week training program. Scientists will administer the program.

AEC cites processing mills for apparent violations

Five uranium processing mills have reportedly received letter orders from the AEC directing them to correct apparent violations of the Commission's radiation safety regulations. The apparent violations involve technical personnel, overexposures and failures to conduct surveys. The AEC has directed each company to submit a complete report on the measures taken to comply with the regulations. In addition, monthly reports beginning at the first of the year will be required with respect to their radiation protection program.

Congress approves AEC transfer of regulatory authority to states

Legislation permitting the AEC to transfer regulatory authority to qualified state governments has been approved by Congress. The states now exercise authority over X-rays and radium applications; the new act adds radioisotopes, source materials (uranium ores, etc.) and special nuclear materials in quantities *not* sufficient to form a critical mass. Regulations are being formulated opening the way for transfer agreements with the state bodies adequately staffed to handle the new responsibilities.

Radiation protection measures agreed on by OEEC countries

The seventeen countries forming the Organization of European Economic Cooperation have agreed on common measures for safeguarding their population from the dangers of nuclear radiation. They adopted a decision recommending member countries to ensure adequate health protection for all persons who might be exposed to ionizing radiation. Appropriate measures must also be taken in connection with emergencies or accidents involving such radiation. All countries are to report on legal and administrative measures that they have taken in this field so that the European Nuclear Energy Agency can make a composite survey by the end of the year. The health protection measures should be based on common norms that have been developed by this group. The norms in turn are based on the recommendations of the International Commission of Radiological Protection.

1960 *Health Physics Society site announced*

It was announced recently by the Health Physics Society that their annual convention will be held in Boston in 1960. The meeting will take place during the last week in June at the Statler Hilton Hotel. Preparations are under the direction of Dr. LESLIE SILVERMAN of the Harvard School of Public Health who is chairman of the Arrangements Committee.

Sea burial total recounted

It was recently announced by the AEC General Manager, A. R. LUEDECKE, that the AEC has so far disposed of the following quantities of radioactivity by sea burial:

(a) In the Atlantic, the major disposal areas are 230 miles and 150 miles southeast of Sandy Hook both off the continental shelf at a depth in excess of 1000 fathoms. About 8000 curies plus the reactor structure of the dismantled Seawolf prototype, which is estimated at 33,000 curies induced in stainless steel, have been dumped to date.

(b) In the Pacific, about 14,000 curies has been dumped at a site 48 miles west of the Golden Gate at a depth of over 1000 fathoms.

Radioisotope directory published

The first volume of an International Directory of Radioisotopes has been published. It contains information on unprocessed and processed radioactive preparations and specific radiation sources. The second volume will contain information on C^{14}, S^{35}, H^{3} and I^{131} and will follow shortly. Also contained in the first volume are all radioisotopes which are for sale or distributed by fifty major suppliers in the world.

Senate committee reports effects of hypothetical nuclear war

A "summary-analysis" of hearings on biological and environmental effects of a nuclear war was released by the Joint Congressional Committee on Atomic Energy setting forth the probable physical and biological effects of such a war on man and his environment. All weapons were arbitrarily designated as having a yield of 50 per cent fission and 50 per cent fusion. A total yield of 1446 megatons were detonated within the United States on 224 targets. An additional 2500 megatons were assumed to have been detonated in attacks on overseas bases. The testimony indicated that such an attack would have cost the lives of 50 million Americans with an additional 20 million sustaining injuries. More than a quarter of the homes would have been destroyed with an additional 13 million homes contaminated by radioactivity. The bursts were designated as surface bursts, 75 per cent of the deaths would be attributed to blast and thermal effects with only 25 per cent due to radioactive fallout. Probably the most significant finding presented was that civil defense preparedness could reduce the radiation casualties of the assumed attack from approximately 30 to 33 per cent. The cost for providing high performance shelter protection for 200 million people was estimated at between 5 billion dollars and 20 billion dollars. The report went on to relate that more than 96 million people in the United States do not live in or near the 224 hypothetical targets and could expect to survive a nuclear attack merely by proper shelter from fallout and a two-week food supply. It was concluded that the United States must have a national radiological defense system and it was emphasized that a nuclear war would not, as many had previously believed, extinguish all human life.

Federal Radiation Council

Following establishment of a Federal Radiation Council, the President directed the Department of Health, Education and Welfare to intensify its radiological health efforts. Having established the central co-ordinating committee, the President directed that the council have *primary* responsibility within the executive branch for analyzing and interpreting data on environmental radiation levels, such as natural background, radiography, isotope and X-ray use, within the medical-industrial sphere. In essence, the new directive entails the relinquishing of the AEC's traditional role as government interpreter and spokesman on fallout. However, according to AEC Chairman, JOHN MCCONE, the AEC will continue to gather data and issue scientific and technical reports on environmental radiation resulting from activities related to the AEC program and will continue its extensive research program to determine the extent of damage to humans from radiation.

Radioactive carbon-14 studies

The C^{14} concentration in the tropospheric CO_2 of the Northern Hemisphere has increased by about 5 per cent per year from March 1955 to March 1958. *During the past four years man has been producing* C^{14} *about fifteen times faster than nature.* The distribution of bomb C^{14} between the hemispheres indicated that horizontal mixing is so rapid that either the troposphere or the stratosphere becomes mixed in less than two years. When the bomb C^{14} produced through March 1958 is distributed uniformly throughout the dynamic reservoir, the concentration of C^{14} in the atmosphere will be 0.25–1.4 per cent higher than in the pre-bomb era.

Gonadal dose from watches and television sets

In two separate surveys, studies were made of the gonadal dose due to luminous dial wrist watches and from television sets.

Dr. S. JOYET of Zurich, Switzerland, reported that the average person wearing a luminous dial wrist watch receives a gonadal dose of 8 mrems/year. The range of gonadal radiation found from "dummied" watches was 4–80 mrems/year.

In a separate study by C. B. BRAESTRAP and R. T. MOONEY of Delafield Hospital, New York, the average yearly gonadal dose from television was found to be 4.3 mrads/year for a viewing distance of 200 cm and 17.2 mrads/year for a viewing distance of 100 cm.

Normal background radiation accounts for a gonadal dose of approximately 100 mrems/year.

Congressional report says current fallout not hazardous but warns against test resumption

The Joint Committee on Atomic Energy released a report warning against resumption of testing at the level of the past five years. They point out that while research efforts have been accelerated in the past two years, the fallout program has not received the high administrative support it needs. It emphasizes that better co-ordination of fallout information is essential, together with the development and application of adequate radiation standards. The analysis points to the need for better co-ordination among *government* agencies in determining, controlling and evaluating environmental hazards.

Testimony was given concerning the following:

While Sr^{90} and Cs^{137} are still considered the greatest hazards, short lived isotopes such as Sr^{89}, Ba^{140}, I^{131} and Zr^{95} are worthy of more consideration.

Long lived C^{14} was described as a *potential* long term hazard from nuclear weapons tests. The danger exists that C^{14} could constitute a genetic hazard comparable to the genetic hazard from other isotopes.

The content of Sr^{90} and Cs^{137} in food has risen even more rapidly than total fallout. Temporarily high levels indicate that under certain conditions Sr^{90} may be taken up directly without going through the soil. Therefore, a consistent degree of discrimination (the selective reduction of Sr^{90} in relation to Ca) from the ground to man cannot be expected.

The concluding fact stated that the biological significance of low levels of radioactivity is still largely *unknown*. The present assumption is that any dose, however small, produces some biological effect and that this effect is harmful.

Film-badge trade group under consideration

A group of commercial film-badge contractors is exploring formation of a new trade association to deal with such common problems as standards, legislation, certification, ethics, insurance and government versus private operation of film-badge services. Response to initial inquiries has been enthusiastic.

Strontium-90 for power

A long lasting electrical power source was predicted using Sr^{90}. Dr. J. G. Morse of Martin Company suggested that radioisotopes be used in power plants for predetermined power output over long periods of time. The idea is to derive useful power through heat generation. An experimental design and development program has been started to yield a completely safe yet reliable power source. This would be capable of producing 100W of electrical energy with a land source efficiency of 5 per cent and marine efficiency of 8 per cent. The central core of Sr^{90} would be protected by a metal container resistant to corrosion, thermal and physical shock and melting.

New radiation hazard to astronautics

Cmdr. Malcolm D. Ross, whose many balloon flights for the Office of Naval Research have made scientific history, recently disclosed that a newly discovered radiation hazard makes manned flight in solar space "appear quite unrealistic for the time being." The new hazard is a result of giant solar flares which send out bursts of radiation and which as yet are unpredictable. This hazard is, in addition to the Van Allen radiation belts, recently discovered by earth satellites and moon probes. Ross added that the dangers of the Van Allen belts "seem pale by comparison."

Measurements made by University of Minnesota scientists indicate that these solar flares might cause a dose rate of 1000 rems/hr to astronautics. Even at 80,000–90,000 ft, radiation intensity during flares amounts to perhaps 10 rems/hr. No information was made available as to the frequency of the flares or to the duration of the hazard due to a given flare.

Health Physics Society

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W. S. SNYDER (Editor, *Health Physics*)
J. J. FITZGERALD (News Editor, *Health Physics*)

Maximum Permissible Body Burdens and Maximum Permissible Concentrations of Radionuclides in Air and Water for Occupational Exposure

NATIONAL Bureau of Standards Handbook 69 (supersedes NBS Handbook 52), 95 pages, 35 cents, issued June 5, 1959. (Order from Superintendent of Documents, U.S. Government Printing Office, Washington 25, D.C.)

This Handbook contains the latest recommendations of the National Committee on Radiation Protection and Measurements (NCRP) concerning the maximum permissible internal exposure of the human body to radioisotopes. These recommendations represent a revision of those published by the NCRP in 1953.

The 1953 recommendations were for occupational exposure, 24 hr/day, continuously for 70 years. Prepared by the Subcommittee on Permissible Internal Dose, the study took five years to complete and included nearly 75 radioisotopes.

The work has continued at an accelerated pace

since 1953 with the added co-operation of the International Commission on Radiological Protection (ICRP). The present Handbook includes maximum permissible concentrations (MPC) and body burdens for some 240 radioisotopes and includes many refinements based on knowledge not available in 1953. The study has included the evaluation of nearly 2000 separate researches reported in the literature, having bearing on the permissible-dose problem.

The new values reflect, where applicable, the lowering of the basic maximum permissible dose (MPD) recommended by the NCRP in 1957 and 1958. As a result of all these considerations, some values have been increased, some decreased, and others remain essentially the same. In addition, the exposure period has been changed from 70 years continuous occupational exposure to 50 years continuous occupational exposure, a more realistic figure for radiation workers. All maximum permissible concentrations (MPC) are given for both 40- and 168-hr weeks.

The recommendations in this Handbook are to be considered only as recommendations of a group of experts in the radiological protection field. They have no legal force requiring or demanding adoption.

Note: Foreign remittances must be in U.S. exchange and should include an additional one-fourth of the publication price to cover mailing costs.

U. S. Department of Commerce
National Bureau of Standards
Washington 25, D. C.

Obituary

Dr. SIMEON T. CANTRIL, member of the Honorary Editorial Advisory Board for *Health Physics* died Thursday, September 10, 1959, of a heart attack at the age of 50. His valuable services will be missed by all in the field of Radiation Protection. Dr. CANTRIL, an expert on the effects of atomic radiation, was director of the Swedish Hospital's Tumor Institute at Seattle, Washington. Before going there in 1938, he was on the staff of the Curie Foundation in Paris. During World War II, he was drafted to take part in the Manhattan Atomic Project and was at the University of Chicago when nuclear fission was first accomplished there. Dr. CANTRIL was Medical Director at the Oak Ridge National Laboratory, Oak Ridge, Tennessee, and later was Assistant Medical Director at the Hanford Plutonium Works. He was also chairman of the Atomic Energy Commissions's Advisory Committee on Biology and Medicine.

Health Physics Pergamon Press 1960. Vol. 2, p. 316. Printed in Northern Ireland

Erratum

THE two figures below were omitted in error from the Note "Some Unusual X-Radiation Dosimetry Problems associated with Radar Installations" by R. ZENDLE and E. E. GOODALE which appeared in Vol. 2, No. 1, p.78.

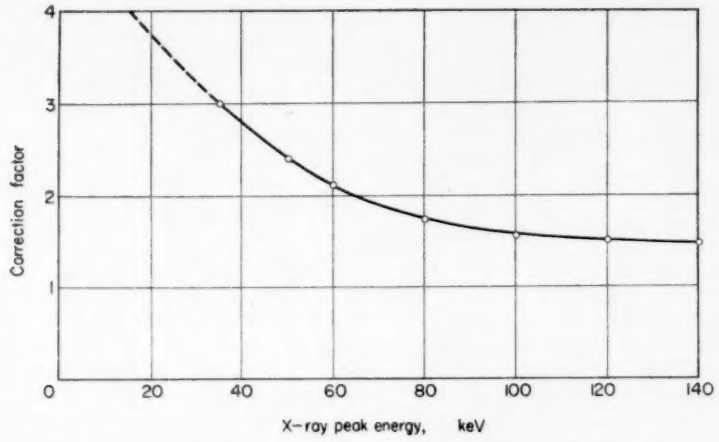

FIG. 1. Correction curve—80 mesh copper-screened ionization chamber.

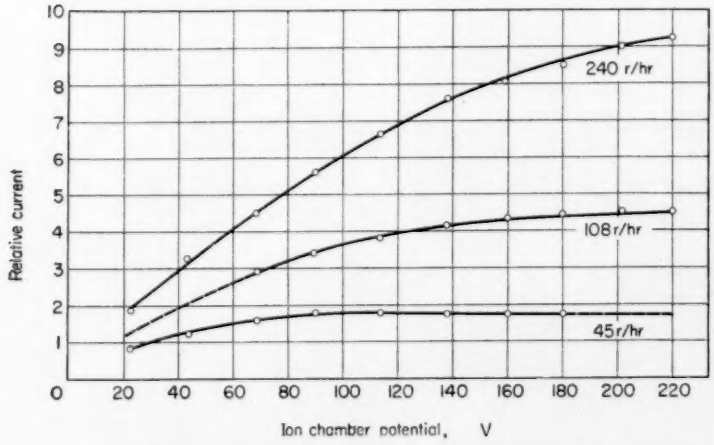

FIG. 2. Saturation curves—high dose-rate exposed ion chamber.

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ADDENDUM TO
ICRP PUBLICATION 1 (1958 RECOMMENDATIONS)

REPORT ON DECISIONS AT THE 1959 MEETING OF THE INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION (ICRP)

INTRODUCTION

THE period since the Commission's last regular meeting in 1956 has been the most active in its history. Because of rapid developments in the field of nuclear energy and the more widespread use of radiation sources of all types, the scope of the work has increased beyond the field of medical radiology into all fields of radiation protection, including industrial uses as well as the exposure of the general public.

In 1955 the United Nations established a Scientific Committee on the Effects of Atomic Radiation to collect and evaluate information on radiation exposure and radiation effects. In addition, bodies such as the World Health Organization, the International Labour Office, the Food and Agriculture Organization and the International Atomic Energy Agency are actively interested in radiation protection problems and look to the International Commission on Radiological Protection for guidance. Although the primary responsibility of the Commission has been to the radiological profession, it has had to widen its scope and has accordingly been active not only during the last two international Congresses of Radiology but also in the intervening period.

Amendments to the 1953[1] Recommendations were reported to the Mexico Congress and published in 1957.[2] Revised Recommendations of the Main Commission were adopted in 1958 and printed early in 1959.[3] Reports of the various Committees exist at present in various stages of preparation. The Report of Committee II on internal radioactive substances is already in press.[4] A revised version of the Committee III Report, which is essentially a code of practice for the radiological profession, was approved during the 1959 meeting and will be published in the near future.[5] The Report of Committee IV on high-energy and heavy-particle radiation is undergoing a final revision. Committee V has prepared a report on the disposal of radioactive waste from hospitals and laboratories and is proceeding as rapidly as possible to prepare a further report on the disposal of waste from atomic energy establishments.

The Commission has an official relationship with the World Health Organization and the International Atomic Energy Agency. There has been close co-operation with the United Nations Scientific Committee on the Effects of Atomic Radiation, and the Commission, jointly with the International Commission on Radiological Units and Measurements, has on two occasions accepted its invitation to perform special studies. The results of the first study on the evaluation of gonad dose from medical procedures were published in 1957.[6] The second study, on the evaluation of exposure relevant to somatic damage, has been initiated and a report is to be prepared before the end of 1960.

The Commission also co-operates with the International Labour Office and the Food and Agriculture Organization and has invited them to send observers to future meetings. The Commission has been invited to send observers to appropriate meetings of the International Organization for Standardization and the United Nations Educational, Scientific and Cultural Organization.

During the 1959 meeting, the Commission discussed its basic Recommendations[3] and although no substantial changes were made, a number of explanatory statements were drafted. These statements which will be presented in the following text include such subjects as the irradiation of pregnant women, the maximum

doses to individuals in the population at large, the interpretation to be placed on the paragraph dealing with the contamination of public air and water supplies, and the hours of work and vacation of radiation workers.

A grant from the Rockefeller Foundation made it possible for the Commission to invite a number of non-member experts on dose-effect relations to participate in the Munich discussions and be available for consultation.

During the preparation of this statement the ICRP has had the following composition.

1956–1959
(Including the Munich sessions)

R. M. SIEVERT, Chairman (Sweden)
G. FAILLA, Vice-Chairman (U.S.A.)
W. BINKS, Secretary* (Great Britain)
L. BUGNARD (France)
H. HOLTHUSEN (Germany)
J. C. JACOBSEN (Denmark)
R. G. JAEGER (Germany)
W. V. MAYNEORD (Great Britain)
K. Z. MORGAN (U.S.A.)
R. S. STONE (U.S.A.)
L. S. TAYLOR (U.S.A.)
E. A. WATKINSON (Canada)
Sir ERNEST ROCK CARLING, Chairman emeritus (Great Britain)

1959–
(Present composition)

R. M. SIEVERT, Chairman (Sweden)
E. E. POCHIN, Vice-Chairman (Great Britain)
W. BINKS (Great Britain)
L. BUGNARD (France)
H. HOLTHUSEN (Germany)
J. C. JACOBSEN (Denmark)
R. G. JAEGER (Germany)
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L. S. TAYLOR (U.S.A.)
E. A. WATKINSON (Canada)
Sir ERNEST ROCK CARLING, Chairman Emeritus (Great Britain)
G. FAILLA, Vice-Chairman emeritus (U.S.A.)
B. LINDELL, Secretary (Sweden)

* Mr. BINKS resigned as Secretary in 1957, for health reasons. After his resignation E. E. SMITH (Great Britain) served as Acting Secretary, and since August 1, 1957, B. LINDELL (Sweden) has served as Temporary Secretary.

EXPLANATORY STATEMENTS AND AMENDMENTS TO THE 1958 RECOMMENDATIONS

In the 1958[3] Recommendations all maximum permissible doses are expressed in rems without reference to the appropriate RBE values. Such reference will be given in the reports of the Committees concerning the application of the recommendations in various specified fields. The RBE values used in each of the Committee reports to be published are consistent with those published in 1955.[1]

The following statements refer to particular paragraphs in the 1958 Recommendations:

Explanatory statement of paragraph 46 (*see also paragraphs* 35 *and* 41)

It has been recommended that "medical exposure" be excluded from the calculation of the maximum permissible dose of those occupationally exposed. The Commission wishes to emphasize that "medical exposure" refers to the exposure of *patients* that is necessary for medical purposes and *not* to the exposure of the personnel conducting such procedure.

Interpretation of paragraph 49 (*see also paragraphs* 10, 14 *and* 52)

A calendar 13-week period may be used instead of a period of 13 consecutive weeks *if there is no reason to suppose that doses are being accumulated at grossly irregular rates.*

Addition to paragraph 49

The following words should be added at the end of the paragraph—"*especially in the case of women of reproductive age.*"

New paragraph to be added after paragraph 51e

"(51f) Where work involves exposure to β-rays of $E_{max} > 2.5$ MeV, eye shields or other suitable shielding may be necessary to keep the dose in the lens within permissible limits. In the case of exposure to β-rays of lower energy, if the provision of such shielding is impracticable, the small additional β-ray dose in the lens over the dose already permitted for more penetrating radiations, such as γ-rays or neutrons, is permissible, provided the dose in the skin is

limited to the level recommended in paragraph 52a."

Additional paragraphs regarding the application of paragraphs 49, 51d *and* 51e *to cases of internal exposure*

With reference to paragraph 49:

"(52e) One or more short-term exposures to radioactive materials (together with any exposure to external radiation) within a period of 13 consecutive weeks is considered acceptable if the total intake of radioactive material during this period does not exceed the amount that would result from intake for 13 weeks at the maximum levels for occupational exposure to such radioactive materials permitted by the 1959 Report of Committee II. The dose to the critical organ during the following 50 years resulting from such an intake will not exceed the quarterly limit on dose stated in the 1958 Recommendations of ICRP. These limits are:

(1) for the whole body and the gonads: 3 rems,
(2) for the skin, thyroid and bone*: 8 rems,
(3) for other organs: 4 rems."

With reference to paragraph 51d:

"(52f) In the case of an accidental high exposure to radioactive materials where the total intake of radioactive material exceeds the amount that would result from intake for one year at the maximum levels for occupational exposure to such radioactive materials permitted by the 1959 Report of Committee II, an estimate of the intake resulting from the exposure shall be entered on the individual's record and he shall be referred to competent medical authorities for appropriate action."

With reference to paragraph 51e:

"(52g) Emergency work involving exposure above permissible limits to radioactive materials shall be planned on the basis that the total intake of radioactive material during the emergency period should not exceed the amount that would result from intake for one year at the maximum levels for occupational exposure to such radioactive materials permitted by the 1959 Report of Committee II. The dose to the critical organs during the following 50 years resulting from such an intake will not exceed the maximum yearly limit on dose stated in the 1958 Recommendations of ICRP. These limits are:

(1) for the whole body and the gonads: 12 rems,
(2) for the skin, thyroid and bone*: 30 rems,
(3) for other organs: 15 rems.

When such an exposure has occurred, an estimate of the intake shall be entered in the individual's record and measures shall be taken to prevent further exposure during the period of time it would take to accumulate this intake at the constant level of occupational exposure permitted by the 1959 Report of Committee II for such materials."

Explanatory statement of paragraphs 53 *to* 57 (*see also paragraphs* 15, 16, 38 *and* 39)

The size of a "group" or a "population" is a relative concept and it is often difficult to decide whether a group should be considered "large" and whether relatively small national populations should be referred to as the "whole" population with respect to exposures of which the importance depends on the size of the group exposed. The size of a group is irrelevant in the case of *individual* exposure, where groups are defined only to distinguish between individuals for whom the recommended maximum permissible doses are different for various reasons or for whom the membership of a certain group provides for more specific recommendations with regard to individual monitoring, medical examinations and surveys of the protection conditions. In the case of, for example, average exposure, however, the "*whole*" *population* is involved and the responsibility for the interpretation of this concept will rest with national and international administrative authorities responsible for the legal application of protection requirements.

Additional section regarding exposure of individual members of the population at large (*to follow after paragraph* 57)

A new section, under the heading "EXPOSURE OF THE POPULATION AT LARGE" should be added

* Based on body burden of 0·1 μc of Ra^{226} (see Report of Committee II).

after the section headed "EXPOSURE OF SPECIAL GROUPS" (paragraphs 53–57). The text to be added, referring to both external and internal exposure, reads: "The maximum total dose limit for *individuals* in the population at large (excluding those occupationally exposed and the special groups B (a) and B (b)) should be that recommended for members of the special group B (c) (cf. paragraph 55)."

Correction to paragraph 65

The figure "1.7" in paragraph 65, Addendum (c), is a misprint for "0.7".

Interpretation of paragraph 68

The basis for the limits of permissible exposure of populations to man-made sources of ionizing radiations is the dose received by the various organs of the body and not the MPC-values, or other criteria by which the dose is controlled. Nevertheless, for planning purposes some guidance as given in paragraph 68 must be available. The word "average" in paragraph 68 refers to the concentration of radioactive nuclides, averaged over a year, in the total intake to the average person of the population.

STATEMENTS ON ITEMS NOT INCLUDED IN THE 1958 RECOMMENDATIONS

Occupational exposure of pregnant women

With regard to occupational exposure of pregnant women the Commission notes that:

(1) It is especially in respect of somatic damage in foetal tissues that pregnant women present a "special-risk problem" in case of occupational exposure.

(2) Any special recommendations for pregnant women must in practice apply to all women of reproductive age.

Occupational working hours and length of vacation

The Commission considers that with the present maximum permissible exposure levels no special treatment of radiation workers with respect to working hours and length of vacation is required.

Emergency exposure of environmental populations

This subject was discussed extensively during the Munich meetings and the Commission considers that the British Report,[7] recommending criteria for acceptable levels under emergency conditions of I^{131}, Sr^{89}, Sr^{90} and Cs^{137} ingested in food or milk, constitutes a useful and sound approach to the subject.

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SPATIAL DISTRIBUTION OF ENERGY DISSIPATED BY FALLOUT β-RAYS

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(*Received* 2 *December* 1959)

Abstract—Calculations are described of the spatial distribution of energy dissipated in air by the delayed beta rays from products of slow neutron U^{235} fission. Results are given for both plane isotropic and point isotropic sources for times after fission of 1.12 and 23.8 hr.

1. INTRODUCTION

THE purpose of this paper is to report calculations of the spatial distribution of energy dissipation in air by the delayed β-rays from products of slow neutron U^{235} fission. In particular, calculations were made to determine the rate of β-energy dissipation at specified distances from point and plane isotropic sources. The input data used were of two types describing (1) the spatial distribution of energy dissipated by *monoenergetic* electrons, and (2) the spectrum of β-rays emitted from U^{235} fission product sources as a function of time after fission.

A theoretical determination of electron energy dissipation functions utilizing multiple scattering theory has been published by SPENCER[1]. This theory is utilized in recent tabulations for monoenergetic electrons in a variety of materials;[2] and these data are the basis for the calculations reported here.†

As far as the writers know, there have been no direct measurements of β-ray spectra for U^{235} fission-product sources. However, such spectra have been determined indirectly for various times after fission by NELMS and COOPER[12]. Their calculations were a synthesis of (1) tabulations by BOLLES and BALLOU[13] of radionuclide, time-dependent activities, and (2) tabulations by MILLER[14] of maximum β-energies for the radionuclides listed by BOLLES and BALLOU. The spectral data for this paper were taken from NELMS and COOPER's calculations.

II. THEORY

Referring to the geometry of Fig. 1, $H(\rho, t)d\rho$ is defined to be the energy dissipated in a spherical shell of radius ρ and thickness $d\rho$ by β-rays emitted at time t after fission by a point source located at P, the center. A related quantity of comparable importance is $I(z, t)dz$, the energy dissipated in plane layers of thickness dz and distance z from the source. It is obtained from $H(\rho, t)$ by integration, i,e.:

$$I(z, t) = \frac{1}{2}\int_z^\infty \frac{H(\rho, t)}{\rho}\, d\rho \qquad (1)$$

$I(z, t)$ also gives energy dissipation as a function of distance from a plane isotropic source.

The function $H(\rho, t)$ may be represented by the integral:

$$H(\rho, t) = \int_0^\infty K(E_0, \rho)S(E_0, t)\, dE_0 \qquad (2)$$

* Guest workers at National Bureau of Standards. Supported in part by O.C.D.M.

† SPENCER's calculations make use of tabulations of stopping power and electron ranges by NELMS[3] and elastic scattering cross-sections by DOGGETT and SPENCER[4]. His computations take accurate account of deflections but neglect straggling in range due to knock-on collisions with atomic electrons.

Earlier efforts to determine β-ray energy dissipation functions have been largely empirical. LOEVINGER made measurements using isotropic plane sources and attempted to fit exponential functions to the resulting data.[5] Additional experimental work by CLARK *et al.*[6] in Chicago, SOMMERMEYER and WAECHTER[7,8] in Germany, and EMERY[9] in England produced consistent results for energy dissipation in air by β-rays from a variety of isotropic point sources. LOEVINGER[10] then applied his curve-fitting techniques to these three sets of data and derived an exponential function representing approximately the point source, β-ray energy dissipation function.

Other theoretical work on β-energy dissipation has been reported by ROESCH[11]. He approximated the dissipation function by an application of the age diffusion theory.

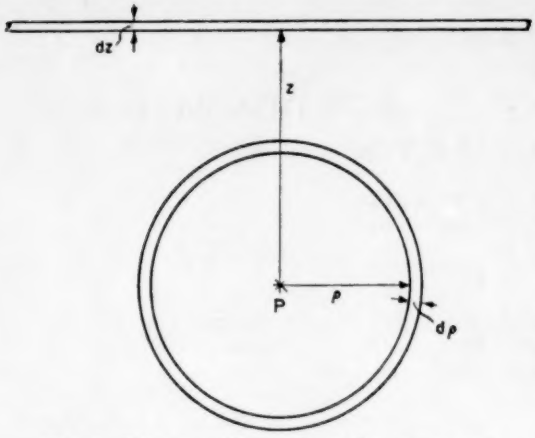

FIG. 1. Diagram illustrating the geometry assumed in the calculations. A point source of β-radiation at P is shown in spatial relationship to a differential spherical shell and a differential slab of the absorbing medium.

where $S(E_0,t)\,dE_0$ is the number of β-rays emitted by the source at time t after fission with initial kinetic energies between E_0 and $E_0 + dE_0$.

The spectral data for times of 1.12 and 23.8 hr were taken from Table 11 of Ref. 12. They are shown in Fig. 2 as energy spectra normalized to correspond to 1 MeV total spectral energy. The quantity $K(E_0, \rho)d\rho$ is the energy dissipated in a spherical shell of radius ρ and thickness $d\rho$ by an electron of initial energy E_0. SPENCER's energy dissipation data were not tabulated directly as $K(E_0, \rho)$, but were instead tabulated in terms of: (1) $(-dE/dx)_0$, the stopping power evaluated at the initial energy; (2) a dimensionless function $J(E_0, \tau)$; and (3) the maximum residual electron range $r_0(E_0)$, where $r_0(E_0)$ is given by the integral:

$$r_0(E_0) = \int_0^{E_0} dE \Big/ \left(-\frac{dE}{dx}\right) \tag{3}$$

The interrelationships between these functions and $K(E_0, \rho)$ are as follows:

$$K(E_0, \rho) = \left(-\frac{dE}{dx}\right)_0 J(E_0, \tau) \tag{4}$$

$$\tau = \frac{\rho}{r_0(E_0)} \tag{5}$$

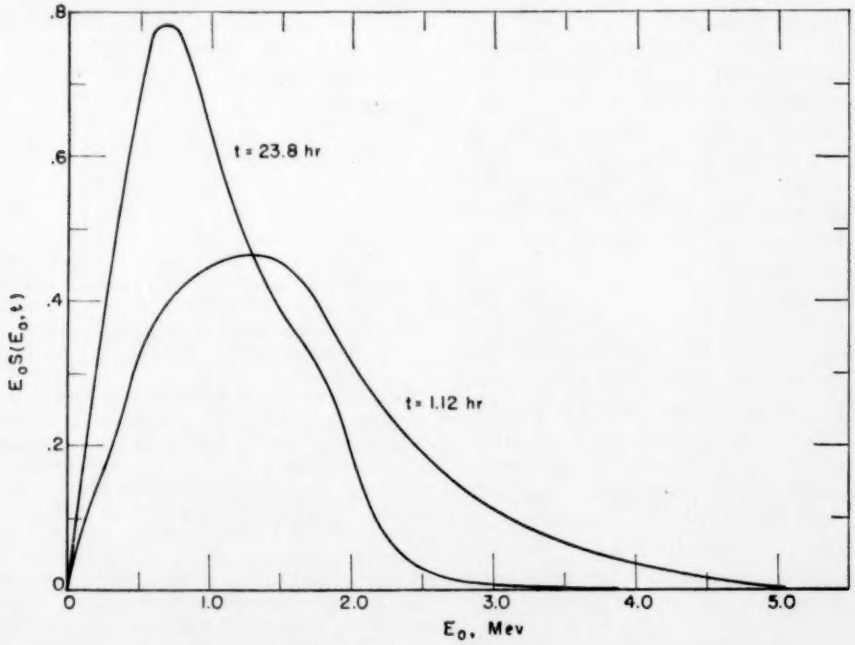

FIG. 2. β-Energy spectra for U^{235} fission product with volatile constituents removed, for times after fission of 1.12 and 23.8 hr.[12] Both curves have been normalized to correspond to 1 MeV total spectral energy.

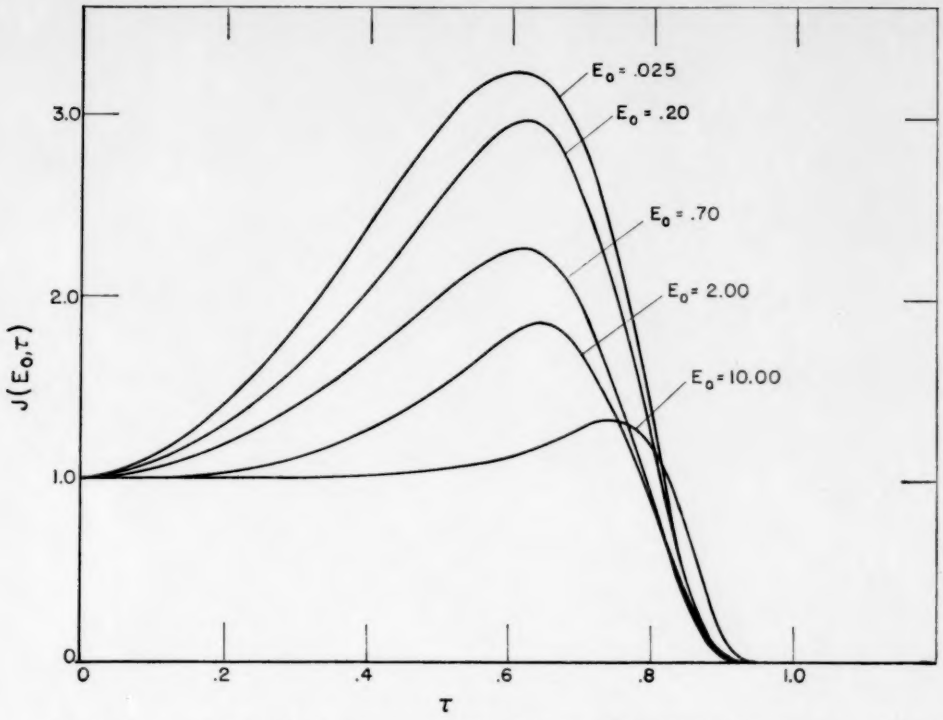

FIG. 3. Curves of $J(E_0, \tau)$, the monoenergetic β-energy dissipation function, vs. τ, the scaled β-ray penetration distance, for representative values of initial electron energy E_0.[9]

Fig. 3 shows graphs of $J(E_0, \tau)$ for several values of E_0.

It was convenient to change the variable of integration in (2) from E_0 to τ since the energy dissipation data were tabulated as a function of τ. This change was accomplished using (4) and (5) to obtain:*

$$H(\rho, t) = \rho^{-1}\int_0^1 \left[r_0(E_0)\left(-\frac{dE}{dx}\right)_0\right]^2 \times S(E_0, t)J(E_0, \tau)d\tau \quad (6)$$

where E_0 in the integrand is determined from (5) as an implicit function of τ.

Numerical evaluation of (6) required interpolation for most values of the integrand. The interpolations were made by plotting graphs of the factors in the integrand and reading the desired values from the curves. Then, for fixed ρ, the integrand was tabulated and the integral evaluated by SIMPSON's rule.

III. DISCUSSION OF RESULTS

Fig. 4 shows the results of both point source and plane source calculations for the times considered. Distance from the source is given in grammes per square centimeter across the bottom of the figure and in feet of air across the top. Each function has been normalized to correspond to unit total energy dissipated.

Cumulative integrals over the functions in Fig. 4 are shown in Fig. 5. The abscissa gives the penetration up to which a specified fraction of the total energy has been dissipated.

Although Figs. 4 and 5 relate to penetration in air, by rescaling they can be fairly accurately applied to water and many organic substances. The important parameter is the ratio of the electron range in the substance to the range in air. This ratio is nearly energy independent,

* This expression is essentially equation (43) of Ref. 1.

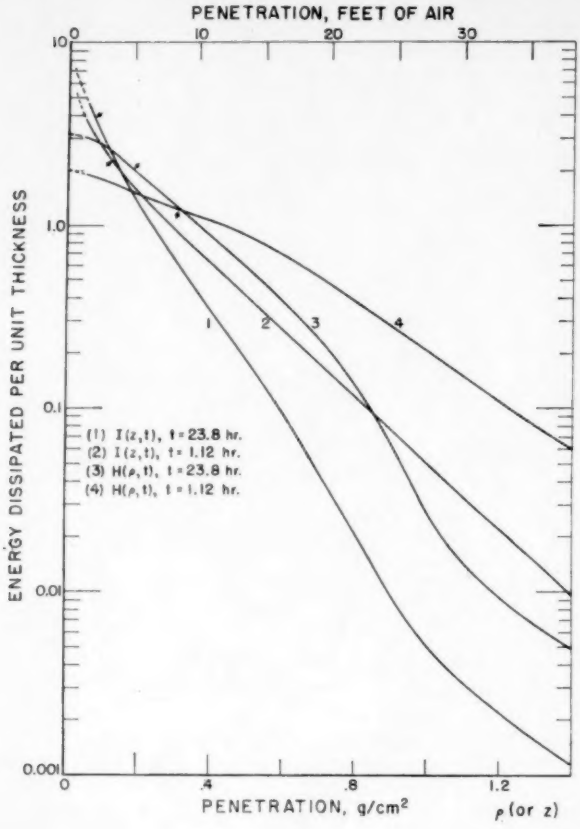

FIG. 4. Spatial distribution of energy dissipation. The curves are all normalized to correspond to unit total energy dissipated.

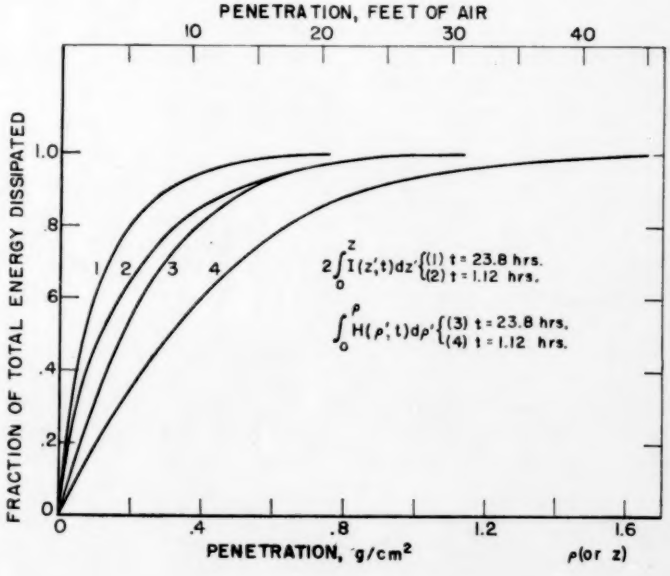

FIG. 5. Curves giving the fraction of total energy dissipated out to a given sphere radius or layer thickness.

and in the case of water and air is approximately 0.87 (at 1 MeV). $H_{\text{water}}(\rho', t)$ normalized to unit total energy dissipated, is given by:

$$H_{\text{water}}(\rho', t) = 1/0.87\, H_{\text{air}}(\rho'/0.87, t) \quad (7)$$

where $H_{\text{air}}(\rho'/0.87, t)$ is given in Fig. 4. The rescaling procedure for other low-Z materials can be carried out in a similar manner; and a rough approximation may be obtained even without rescaling.

The arrows of Fig. 4 locate the depth of penetration at which 50 per cent of the total input energy is dissipated. At 1.12 hr for the point-source function, the 50 per cent penetration occurs at 0.31 g/cm^2, while at 23.8 hr, this point occurs at 0.18 g/cm^2. This tendency for the radiation to be softer at the longer time after fission is also reflected in the plane source functions; e.g. at 1.12 hr, half the total energy is dissipated at a penetration of 0.12 g/cm^2, while at 23.8 hr, the 50 per cent dissipation mark occurs at 0.07 g/cm^2. All penetration curves, and spectra of Fig. 2 show clearly that β-rays are much less penetrating at the later time.

The effectiveness of air as a protection against β-radiation can be seen by noting that at a time of 1.12 hr after fission half the β-energy emitted from ground contamination is absorbed within a few feet of air.

During the course of these calculations the point and plane sources have been assumed to be, respectively, infinitely small or infinitely thin, i.e. attenuation due to self-absorption by the source has been neglected. However, if one assumes a typical fallout particle to be 200 μ in diameter with density comparable to that of sand,[15] the effective penetration depth of the source its iself approximately 0.02 g/cm^2. A glance at Fig. 5 indicates that this is equivalent to an initial β-attenuation of from 5 to 20 per cent depending upon the geometry and time after fission.

IV. CONCLUDING REMARKS

The work described in the paper is intended as a pilot project for more extensive calculations involving other fission spectra and β-absorbers, and other β-emitters.

Acknowledgment—The authors wish to acknowledge their indebtedness to Dr. L. V. Spencer for his guidance during the course of this work.

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PLUTONIUM ACCUMULATION FROM LONG-TERM OCCUPATIONAL EXPOSURE*

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Abstract—Analyses of tissue aliquots from a plutonium process operator, who had been exposed to Pu^{239} largely via chronic low-level inhalation for approximately 6 out of $11\frac{1}{2}$ years of employment, showed that he had accumulated a body burden of approximately 0.018 μc. Estimations of his body burden from his urine assay record ranged from 0.019 to 0.034 μc. The highest plutonium concentration (125 dis/min per g) was found in pulmonary lymph nodes, followed by liver (9.9 dis/min per g), lungs (4.8 dis/min per g) and bone (average of sternum, rib and vertebra, 1.4 dis/min per g). Some implications of these findings to chronic low-level inhalation exposures and to estimation of body burden from urine assays are discussed.

THE occurrence of a fatal criticality accident at the Los Alamos Scientific Laboratory provided an opportunity to obtain data on plutonium accumulation in an individual who had spent a total of approximately 6 years of an $11\frac{1}{2}$-year employment period working with this potentially hazardous material.

These data are of interest because they provide, for the first time, limited information on (a) the efficacy of industrial hygiene and engineering practices in plutonium processing operations; (b) body distribution of plutonium in an individual, exposed primarily by inhalation of low-level plutonium aerosols over a prolonged period; and (c) reliability of estimates of body burden from urinary excretion data, when exposure has been primarily via inhalation.

The individual involved was a 75-kg male, 38 years of age at the time of death. The fatal accident occurred during a plutonium recovery procedure. Details of the accident and the operation are described elsewhere.[1] Death resulted from an over-dose of radiation. No plutonium contamination occurred and, consequently, the conditions of the accident did not influence the findings in this study.

The individual's employment and job assignment history is shown in Table 1.

Table 1. *Employment history of the subject*

Period	Job assignment	Time (days)
17 June 1946—24 January 1949	Pu^{239} recovery operator	947†
24 January 1949—4 June 1954	U^{235} recovery operator	1987
4 June 1954—8 June 1955	Resigned—no exposure	369
8 June 1955—31 December 1958	Pu^{239} recovery operator	1297†
Total plutonium exposure time		2244

* Work performed under the auspices of the U.S. Atomic Energy Commission.
† Between first and second plutonium exposure periods, exposure conditions had been greatly improved.

Table 2. Average concentration of airborne plutonium in areas in which employee was assigned

Year	Room 308 (dis/min per m^3)	Room 313 (dis/min per m^3)	Room 408 (dis/min per m^3)	Room 413 (dis/min per m^3)	Rooms 212, 213, 218 (dis/min per m^3)
1946	6	16	188	15	
1947	11	25	77	98	
1948	Not used	24	35	69	
1949	Not used	19	30	72	
1949–1954	Worked with enriched uranium				
1955					3
1956					3
1957					4
1958					4

During the first period of plutonium exposure (17 June 1946—24 January 1949), his work consisted of chemical processing involving plutonium nitrate solutions, plutonium oxalate and, occasionally, plutonium fluorination. During the second period (8 June 1955—31 December 1958), his work consisted largely of liquid–liquid extraction of plutonium under greatly improved exposure conditions.

Detailed exposure records were kept during the periods when the employee was working with plutonium. These records included plutonium air concentrations in the processing rooms, the operator's nose swipe counts done several times a week up to the end of 1955 and frequently but irregularly thereafter, daily hand counts, and frequent plutonium assays of 24-hr urine specimens. Average plutonium air concentrations to which the individual was exposed are presented in Table 2. The instances when his nose swipe counts went above 50 dis/min (arbitrarily chosen as the limit of significance) are listed in Table 3, and his urine assay record is given in Table 4. No tabulation of hand counts is given, since they were consistently below significant levels. Results of fecal analyses were not available. Because of the analytical and sampling difficulties involved, fecal analyses are not a routine practice at the Los Alamos Scientific Laboratory.

There were no specific accidents to which the individual's exposure could be attributed. However, as might be expected, a number of

*Table 3. Record of high nose swipe counts**

Date		Right/Left nostril (counts/min)
1946	29 July	189/320
	30 July	87/70
	5 September	149/19
	25 September	55/4
	1 November	61/15
	10 December	57/68
	30 December	164/106
1947	21 January	102/61
	1 April	91/135
	7 July	144/40
	3 October	120/78
1948	10 February	0/59
	26 April	86/3
	9 June	33/50
	10 June	244/72
	2 July	72/1
	2 August	65/0
	1 December	50/38
After 1948 (none)		

* Technique consisted of rotating a piece of dampened filter paper (on an applicator) in each nostril, after which the paper was unrolled, flattened, dried and counted. Fifty counts/min was arbitrarily taken as the limit of significance.

Table 4. Results of periodic plutonium urine assays for entire period of employment

Date	dis/min per 24 hr specimen	Date	dis/min per 24 hr specimen
9 August 1946	1.2	1 August 1955	1.2
19 September 1946	4.7	12 August 1955	0.6
18 December 1946	1.7	19 August 1955	0.0
18 April 1947	0.7	30 September 1955	0.7
23 May 1947	0.7	14 November 1955	0.7
26 June 1947	0.7	27 December 1955	0.6
30 July 1947	0.0	9 February 1956	0.0
27 August 1947	1.0	5 April 1956	0.7
2 October 1947	1.5	30 April 1956	0.0
7 November 1947	0.8	8 June 1956	0.4
8 December 1947	4.0	20 July 1956	0.6
13 January 1948	1.0	23 August 1956	0.5
13 February 1948	0.0	25 September 1956	0.0
19 March 1948	0.0	24 October 1956	0.0
22 April 1948	2.0	23 November 1956	0.0
23 June 1948	3.7	17 December 1956	0.1
22 July 1948	0.0	31 January 1957	0.23
19 August 1948	0.8	28 February 1957	0.68
20 September 1948	3.0	12 April 1957	0.22
26 October 1948	2.0	14 May 1957	0.12
22 November 1948	2.5	14 June 1957	0.11
21 December 1948	0.0	15 July 1957	0.03
24 January 1949	Uranium	19 August 1957	0.00
31 January 1949	2.0	20 September 1957	0.20
14 July 1949	1.2	22 October 1957	0.21
8 February 1950	0.8	31 October 1957	0.39
1 September 1950	0.1	14 November 1957	0.51
28 February 1951	0.8	10 January 1958	0.00
4 September 1951	0.7	21 February 1958	0.65
19 May 1952	0.3	25 March 1958	0.51
14 December 1952	0.0	7 May 1958	0.25
4 September 1953	0.0	19 June 1958	0.55
4 June 1954	Off uranium	30 July 1958	0.49
18 June 1954	0.0	15 September 1958	0.79
8 June 1955	Back on plutonium	28 November 1958	0.47

minor mishaps occurred during the period of employment. The detailed record of such minor incidents is summarized in Table 5.

At the time of autopsy, tissues were taken specifically for plutonium assay. The specimens taken and their plutonium contents are shown in Table 6. The assays were carried out using the alpha track counting method.[2] Two independent analyses of aliquots from each tissue sample were made by the analytical section of the Los Alamos Industrial Hygiene Group, and aliquots or samples of rib, sternum, vertebrae, lungs, liver and lymph nodes were sent to the Hanford Atomic Products Operation, where they were analyzed independently by both the Biological Laboratory and the Bioassay Group. The values given in Table 6 are averages and standard deviations for the four independent analyses. The standard deviation for the lymph nodes includes variation in

Table 5. Summary of minor exposure incidents during period of employment

Date	Type of incident	Potential exposure
26 August 1946	Minor laceration on thumb	No alpha activity detected
30 December 1946	Helped clean up spill of plutonium solution	Air concentration 52 dis/min per m^3 for 8-hr day; nose count 164/106 dis/min
22 April 1947	Received nitric acid burns on arm	No record of contamination
3 September 1947	Small cut on hand, received while making material transfer	No alpha activity detected
14 December 1947	Puncture wound on right hand from a screw point on an instrument panel	No contamination reported
17 March 1949	Helped clean up spill of enriched uranium	No air concentration or nose swipe data recorded
22 April 1950	Exposed to fumes of oxides of nitrogen	No contamination involved
10 August 1953	Helped clean up spill of enriched uranium	Room air concentration 8 dis/min per m^3; nose count 4 dis/min
9 August 1955	While adding caustic to a plutonium and americium solution, materials sprayed out of reaction vessel. Operator was wearing face shield and respirator	No body contamination detected; nose count 22/28 dis/min; room air concentration 175 dis/min per m^3
25 November 1958	Maintenance work involving removal of plutonium-contaminated insulation from an evaporator–condenser	Nose swipe count 0 dis/min; room air concentration 1070 dis/min per m^3. Operator was wearing Wilson 800 respirator

Table 6. Plutonium concentration in autopsy samples (based on actual analytical data)

Organ or tissue	Gross weight at autopsy (g)	Plutonium concentration (dis/min per g wet wt.)
Psoas muscle		0.01
Kidneys	270	0.05
Heart	400	0.06
Cartilage		0.14
Spleen	116	0.18
Sternum		1.0 ± 0.2
Ribs		1.2 ± 0.3
Vertebrae		2.1 ± 0.6
Lungs (minus major bronchii)	850	4.8 ± 0.6
Liver	1950	9.9 ± 1.4
Bronchial lymph nodes		125 ± 57*

* Includes variation in different lymph-node samples, as well as analytical variation.

sample, as well as variation in analysis, since different lymph-node samples were used in the independent determinations. The highest plutonium concentration was found in the pulmonary lymph nodes (125 ± 57 dis/min per g), followed by the liver (9.9 ± 1.4 dis/min per g), lungs (4.8 ± 0.6 dis/min per g) and then vertebrae (2.1 ± 0.6 dis/min per g). The average of all bone samples was 1.4 ± 0.7 dis/min per g, for which the standard deviation includes the variation among the different bone samples plus the analytical variation.

The total organ and body content, calculated from the data in Table 6, are shown in Table 7. The estimated total body burden was 0.018 μc.

As a result of changes in production methods, the Pu^{238}/Pu^{239} ratio of plutonium processed at the Laboratory increased considerably between 1946–1948 and 1955–1958. Determination of the Pu^{238}/Pu^{239} ratios in bone, lymph node, liver and lung samples indicated bone and lymph nodes had the lowest ratio (corresponding generally to material produced in 1946–1948),

Table 7. Total body burden estimated from tissue or organ weight and plutonium content

Organ or tissue	Weight (g)	Total content (dis/min)
Liver	1950	1.930×10^4
Skeleton (average)	10,000	1.400×10^4
Lungs(minus bronchii)	850	0.408×10^4
Respiratory lymph nodes	10	0.125×10^4
Muscle	30,000	0.030×10^4
Heart	400	0.002×10^4
Spleen	116	0.002×10^4
Kidney	270	0.001×10^4
Balance*	26,400	0.026×10^4
Total	70,000	3.924×10^4 (0.018 μc)

* Assuming plutonium content of the balance is the same as that of muscle.[3]

and lung samples had the highest (corresponding roughly to more recently produced material). The Pu^{238}/Pu^{239} ratio in the liver was intermediate.

DISCUSSION

The data in Tables 2 and 3 show that all high plutonium air concentrations to which the employee was exposed and all high nose swipe counts recorded occurred during his earlier period of exposure. It is very likely that most of his plutonium burden was accumulated during this period. The record summarized in Table 5 shows that there were no specific incidents to which his plutonium exposure could be attributed. The summary is given principally to point out the close attention given to all potential modes of exposure and thereby emphasize the certainty with which a contaminated accident can be ruled out as the source of the subject's plutonium burden. It is most likely that the body burden, in this case, resulted from chronic inhalation exposure to a low-level plutonium contaminated atmosphere. The above speculations regarding time and mode of exposure are supported also by the indication that the Pu^{238}/Pu^{239} ratio in lymph nodes and bone appeared to correspond to that of plutonium being processed during the early period of the subject's exposure.

Three different urine assay procedures were used during the 12-year period over which the urine data shown in Table 4 were collected. Each change resulted in somewhat greater reliability of the data. In 1957, the method was changed to the Hanford alpha track counting procedure.[2] Urine assays from this time onwards have considerably higher reliability than previously. Even during 1957 and 1958, however, there was considerable variation in the assays, which is probably due both to analytical limitations and to normal physiological fluctuations. The employee's systemic plutonium burden was estimated from the urine assays using empirical equations derived from human excretion data.[3] Following an acute exposure occurring at known time, the retained plutonium body burden (D_R) is given by the expression

$$D_R = 435\, U t^{0.76} \qquad (1)$$

in which U is the plutonium (counts/min, dis/min, μc) in a 24-hr urine sample collected t days after the time of exposure. D_R is given in the same units used to express U. Since this equation is applicable to relatively acute exposure occurring at known time, it is necessary in protracted exposure cases to assume an effective time of exposure which, to a first approximation, may be taken as the midpoint of the work period. Following chronic invariant

exposure to plutonium, the total systemic intake (D_t) is given by the expression

$$D_t = \frac{130 \times m \times U}{(n + 1/2)^{0.26} - (n - m + 1/2)^{0.26}} \quad (2)$$

in which m is the duration of exposure (days), and U is the average dis/min per 24-hr urine sample taken n days from the beginning of exposure. D_t is the sytemic exposure and it is rigorously necessary to subtract the amount excreted in order to obtain the amount retained (D_R). However, since only about 10 per cent of a plutonium burden is excreted in 10 years,[3] such a correction is insignificant in most cases. The above expressions indicate the determination of body burden from a single 24-hr urine assay. Analytical limitations of the assay methods and normal physiological variations in urinary plutonium excretion make such estimates completely unreliable, and in practice it is better to use the average of several consecutive assays (even though they may be weeks or months apart).

Because of the nature of the exposure, neither of the above equations is specifically applicable to the case under consideration. Application of equations (1) and (2) to the average of all urine assays run during 1949–51 (during which time there was no exposure to plutonium) gives 0.033 and 0.031 μc, respectively, for the employee's body burden at that time as a result of his first period of exposure. During 1957–58, frequent urine assays were run using the more sensitive and reliable alpha track counting method. Estimations based on equations 1 and 2 (assuming all his exposure occurred during the earlier work period, and using the average of the 1957–58 urine assays) gave 0.034 and 0.031 μc, respectively, for the body burden at the time of death. The latter estimates may be high, since they are predicated on the assumption that the entire body burden was accumulated during the earlier work period and that exposure during the second period made no contribution to the 1957–58 average urine assay value. That this assumption is approximately correct is borne out by the estimates of burden at the end of the employee's first period of exposure. A method of estimating plutonium body burden employing IBM-704 Fortran programming of all urine assay data has been developed by LAWRENCE[4]. His estimate of the subject's body burden at time of death was 0.019 μc. The burden derived from tissue analyses (Tables 6 and 7) was 0.018 μc, which is 45 per cent of the maximum permissible level of 0.04 μc.[5]

The above agreement between body burden from tissue analyses and estimated burden from urine assays is so very satisfactory that it is undoubtedly fortuitous. Determination of body burden from tissue analyses is subjected to considerable sampling uncertainty. Since it is not possible to analyze the entire body, it is necessary to analyze aliquots of the various organs and, on the assumption of uniform distribution and representative sampling, calculate the total body content from the organ weights. This assumption is particularly uncertain with regard to the skeleton, as shown by the analytical data for sternum, rib and vertebra (Table 6).

Some uncertainty in the body burden estimated from urine assays may be expected also from the uncertainty in the exponents of the time parameters in equations (1) and (2). When $t = 10$ days, a 10 per cent error in the exponent of [equation (1)] would make an 18 per cent error in the body burden estimated from the urine assay. The error would increase with time, approaching a factor of about 2 at $t = 3500$ days. Indication that the exponent of t in the basic urinary excretion equation[3] may be in error (by about 10 per cent), in the direction resulting in overestimation of the body burden, was obtained recently by re-estimation of the plutonium burdens of workers exposed at the Laboratory during 1944–45 and who had received no subsequent exposure. When estimated from a urine sample collected approximately 10 years after termination, their systemic burdens were, in most cases, about a factor of 2 higher than the estimates based on urine samples collected during or immediately after exposure. Most of the cases, however, were believed to be inhalation exposures and it is possible, although seemingly unlikely, that their systemic burdens did increase as a result of absorption of plutonium deposited in the lungs and pulmonary lymph nodes.

The distribution of plutonium in the various tissues and organs was somewhat surprising. Approximately 50 per cent of the body burden was found in the liver and 36 per cent in the skeleton (Table 7). A hundred and fifty days after intravenous injection of Pu^{4+}-citrate in man, approximately 65 per cent of the administered dose was found in the skeleton and 22 per cent in the liver.[6] Rat experiments show that the relative concentration of plutonium in bone and liver following intravenous injection is dependent on the chemical form and valence state of the material administered.[7] It is quite possible, in the present case, that the partitioning of plutonium between the liver and skeleton was influenced both by the chemical or physical nature of the plutonium and by the route of exposure. The respiratory route of exposure was undoubtedly responsible for the high plutonium concentrations found in the lungs and pulmonary lymph nodes.

Some interesting qualitative speculations regarding relative deposition and clearance rates of plutonium in various tissues may be drawn from the Pu^{238}/Pu^{239} ratios in bone, lymph nodes, liver and the lungs. The Pu^{238}/Pu^{239} ratios of the plutonium deposited in bone and lymph nodes appeared to correspond with that of plutonium produced in 1946–1948 (which was the subject's earlier and greater period of exposure). The Pu^{238}/Pu^{239} ratio of the material deposited in the lungs, however, appeared to correspond to that of more recent production. The above observations suggest (a) a relatively rapid clearance rate for plutonium in the lungs, compared to that in bone and lymph nodes; and (b) that a relatively small percentage of the material deposited in the lungs must migrate to the latter tissues. Since the Pu^{238}/Pu^{239} ratio of material in the liver was intermediate between that deposited in the bone and lymph nodes and that in the lungs, the rate of clearance from the lungs to the liver must be relatively fast and the retention time in the liver must be longer than in the lungs.

The biological effective dose rate to the various tissues of the subject may be estimated from the observed plutonium concentrations (Table 6) and the following expression derived from the Recommendations of the International Commission on Radiological Protection.[8]

$$R = \frac{C \times \Sigma E(\mathrm{RBE})n}{2.8 \times 10^{-3}}$$

In this expression, R is the dose rate in rems/week, C is the plutonium concentration in μc/g and $\Sigma E(\mathrm{RBE})n$ is an effective energy weighted for the RBE of alpha particles and the nonhomogeneity of energy distribution and absorption in the tissue. The weighted energy terms for plutonium in soft tissue and bone are 53 and 270, respectively. Calculated in the above manner, the pulmonary lymph nodes were subject to the highest biologically effective dose rate ($\sim$1 rem/week), the liver next (0.08 rem/week), followed by the bone and lungs (0.06 and 0.04 rem/week, respectively). The estimated alpha radiation dose to the lymph nodes may be considerably too high because of shrinkage in lymphoid tissue mass as a result of the acute radiation dose ($\sim$10,000 rads of fast neutrons plus gamma rays incident to the chest) received during the criticality accident.

No definite conclusions can be drawn from a single case, but these observations again bring up the troublesome questions of choice of the critical organ following chronic inhalation exposure, the size of the critical tissue volume and the relative sensitivity of various tissues to chronic radiation damage.

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THE DISTRIBUTION AND EXCRETION OF THALLIUM-204 IN THE RAT, WITH SUGGESTED MPC'S AND A BIO-ASSAY PROCEDURE*

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Abstract—The distribution and excretion of thallium in the rat has been studied following injection of Tl^{204} by six routes: intramuscular, intraperitoneal, intratracheal, intravenous, oral and subcutaneous. In all cases absorption is rapid and distribution diffuse with comparatively large quantities found in the kidneys. It is assumed that thallium is generally incorporated into the intracellular fluids. There is no day to day variation in relative organ thallium content with the exception of hair in which there is an actual build-up of thallium with time. Any organ except hair may therefore be said to have the same biological half-time as the body burden (3.3 days) which follows a single exponential function extrapolating to 100 per cent at zero time.

Excretion assumes the same pattern after all routes of injection with some minor exceptions. There is a fecal to urinary ratio of between 2 and 5, a greater rate of decrease in thallium content being found in the urine than in the feces.

A maximum permissible body burden and maximum permissible concentrations for air and water have been calculated for Tl^{204} with kidney as critical organ. Assuming body burden equilibrium at about 30 days exposure on an industrial routine, a suitable method for bio-assay has been suggested.

INTRODUCTION

THALLIUM is used in the manufacture of numerous alloys, of tungsten lamps, of glass with a high refractive index and of rodenticides and insecticides. Medically, it has been used primarily as a depilatory. It is considered to be as dangerous as lead and arsenic[1], and poisoning has occurred resulting from its internal use for ringworm and from accidental and intentional ingestion of commercial poisons. There are no recorded industrial fatalities.[2] The toxicity of thallium may be either chemical in origin, from the stable isotopes Tl^{203} and Tl^{205}, or radiological in nature from the radioactive isotopes. Of these, Tl^{204} would be the most hazardous because of its long half-life and energetic beta particle.

Several studies of the distribution and excretion of thallium have been carried out following oral[3,4], intraperitoneal,[4] subcutaneous[4–6] and intravenous[7] injection. They indicate that thallium is (a) rapidly absorbed by the body, (b) evenly distributed to all tissues except the kidneys in which the concentration of thallium is high and (c) excreted rather slowly, greater amounts being found in the feces than in the urine. In general these studies are not extensive enough to render the establishment of MPC's, and inconsistencies in the findings are quite prevalent. The data of DURBIN *et al.*[8] indicate a rather short biological half time (5.2 days), the value currently in use by the NCRP for calculation of the radiological MPC's for thallium isotopes. This figure is

* This paper is based on work performed under contract with the United States Atomic Energy Commission at the University of Rochester Atomic Energy Project, Rochester, New York.

based on a limited study but agrees closely with that obtained from the results to follow.

The studies to be reported here were performed in order to better establish an effective half-time of thallium in the body, to aid in understanding its behavior in chemical toxicity and to enable calculation of the maximum permissible concentrations for the radioactive isotopes. This was accomplished by comparing the metabolism in rats after six routes of administration; intramuscular (IM), intraperitoneal (IP), intratracheal (IT), intravenous (IV), oral (O) and subcutaneous (SQ). Tl^{204} produced at the Oak Ridge National Laboratory was used in tracer quantities. This isotope decays with a half-life of approximately 3.0 years with a beta emission having an energy of 0.76 MeV.[9] There is no gamma emission associated with Tl^{204} but there is a 70 keV photon from mercury owing to *K*-electron capture. The ratio of the *K*-capture process to the beta emission is about 0.015.[10]

METHOD

Rats were injected with a thallous nitrate solution at a pH of 2–4, and with a specific activity of about 2000 mc/g. For two routes, IP and SQ, the solution was neutralized with potassium hydroxide to a pH of about 6. The volume of solution and concentration of thallium injected varied from route to route.* The rats, all males (University of Rochester strain, Wistar derived, 1923), weighed approximately 200 g. For each route, thirteen animals were injected and of these, four were kept in TUTTLE and BAXTER metabolism cages[11] throughout the experiment. Urine and feces were collected daily and the screening and drainage trays rinsed with N/2 HCl which was then added to the urine samples.

Sacrifice procedures were the same for all routes. Three animals were sacrificed on each of the first, second and seventh days of the experiment and the four metabolism animals on the twenty-first day. The animals were anaesthetized with ether and sacrificed by maximal blood withdrawal. Dissections were made on all animals sacrificed on days 1, 2 and 7, and on two or more animals on the 21st day. Each animal was shaved and skinned and the tail, paws, and salivary glands removed. The abdominal viscera were then removed followed by the thoracic organs. After removing the brain all bones were stripped of muscle as completely as possible. Each organ or tissue was placed in a cellophane bag and the wet weight determined.

The counting was done in one of two Welch Allyn GR Detectors,* counting the 1.5 per cent *K*-capture photons. This method was used to eliminate unnecessary tissue digestions and experimental error due to autoabsorption of the beta particles. The overall counting efficiency was about 0.032 per cent (counter efficiency, 2.1 per cent × *K*-capture, 1.5 per cent). Blood samples were weighed and counted in glass test-tubes. Urine was evaporated to a small volume in a water bath prior to counting, and feces and tissues in cellophane bags were placed in plastic tubes for counting to avoid contamination. The total recovery for all six routes of administration ranged from 87 to nearly 100 per cent of the administered dose.

During the injection of each group, three to five dummies were made by placing an equivalent dose in a test-tube. To arrive at one value for the administered dose in each experiment, a mean dummy value was determined for each counting tube for each day. Because the decay of Tl^{204} over a 21-day period may be considered insignificant, the average of these daily means gave one dose figure for each experiment. The standard deviations of the overall means were then calculated,† the largest deviation being no more than 2.4 per cent of the value taken as the administered dose. Significant counting levels were determined by standard statistical methods for a 95 per cent confidence interval.

RESULTS AND DISCUSSION

The distribution of thallium in tissues and in excreta is presented in tables and graphs to

* Amount injected per rat: IV–20 μc, IM–50 μc, SQ–50 μc, IT–63 μc, IP–75 μc, O–400 μc.

* Welch Allyn High Sensitivity Gamma Radiation Detector, Welch Allyn, Inc., Skaneateles Falls, New York.

† The statistical procedures used throughout this paper were directed by Dr. ARTHUR M. DUTTON of the Statistics Section, University of Rochester Atomic Energy Project.

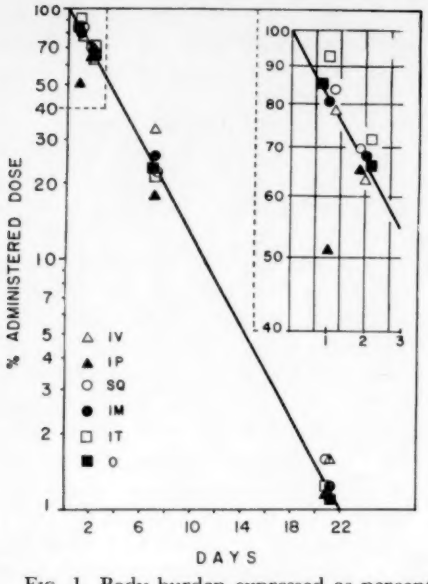

FIG. 1. Body burden expressed as percentage administered dose.

follow. Few 21-day data are shown, as nearly all of the values at that time were insignificant.

Distribution

Fig. 1 shows body burden as percentage administered dose for all routes. Each point represents three animals except at 21 days where each point is an average of four animals. The line was drawn using the median of each group of points. Body clearance occurs exponentially with a half-time of 3.3 days, independent of the route of administration for the first 21 days or until about 1 per cent of the administered dose remains. No measurements were made after 21 days but it may be reasonable to assume the same rate of decrease beyond this point. Some other experimenters previously cited have indicated that some thallium remains in the body for a long period of time.

Tables 1 through 4 show that there is very little day to day or route to route variation in the relative thallium content of any given tissue. On a per organ basis, statistical analysis of variance showed that there may be a significant difference in kidney content between IV and O administration. Day to day differences are most obvious in hair where there is a buildup

Table 1. *Day to day variation of the mean percentage body burden per organ as measured by the standard error. Each value is the mean of all animals from all routes sacrificed on a given day*

Organ or Tissue	1 day	2 days	7 days	S.E.
Muscle	32.66	35.07	33.24	1.21
Bone	20.24	20.61	19.59	0.79
G. I. tract	15.95	11.90	12.14	1.39
Skin	9.08	9.34	10.34	0.69
Kidney	8.13	8.32	7.92	0.40
Liver	4.04	4.35	4.57	0.18
Testes	2.36	3.00	2.85	0.15
Respiratory system	0.99	0.89	0.95	0.17
Salivary glands	0.57	0.59	0.52	0.10
Hair	0.47	0.25	1.56	0.22
Spleen	0.41	0.35	0.37	0.12
Brain	0.38	0.49	0.40	0.05
Heart	0.44	0.41	0.32	0.06

of absolute thallium content with time, such that at 21 days it may contain up to 60 per cent of the body burden. With the exception of kidney and hair, all of the tissue distribution data can be treated as one population. The relative thallium contents of individual organs through 7 days for all routes of administration have been combined in Table 5. The diffuse distribution of thallium is seen, kidney being significantly higher than any other organ on a per gram basis. The salivary glands contain the next highest concentration though markedly less than kidney. This localization of thallium in the salivary glands has been noted by TRUHAUT.[6] Each value represents an average of 54 animals, and the deviations from one to another, as seen in the accompanying standard errors, are very small. Organ weights used in calculating body burden per gram are averages from all animals sacrificed throughout the six experiments.

With the exception of kidney and perhaps one or two other organs, it appears that thallium is incorporated in the intracellular fluids. This was also noted by LUND[5]. The fact that blood (not included in the data shown) contains little

Table 2. Route to route variation of the mean percentage body burden per organ as measured by the standard error. Each value is the mean of all animals sacrificed on 1, 2 and 7 days for a given route of administration

Organ or tissue	IV	IP	IM	SQ	IT	O	S.E.
Muscle	33.52	32.80	35.72	33.70	33.18	33.03	1.71
Bone	20.61	17.63	20.51	20.31	20.30	21.52	1.12
G. I. tract	13.27	14.56	11.27	11.23	14.49	14.06	1.96
Skin	9.71	9.51	9.24	9.88	10.11	9.06	0.97
Kidney	9.72	7.28	8.64	8.72	7.67	6.69	0.57
Liver	4.18	4.63	3.91	4.77	4.12	4.31	0.25
Testes	2.55	2.24	2.95	3.14	2.58	2.97	0.21
Respiratory system	0.94	0.99	1.00	0.95	0.97	0.84	0.12
Salivary glands	0.37	0.60	0.71	0.71	0.36	0.63	0.07
Hair	0.21	0.73	0.78	1.70	0.58	0.58	0.32
Spleen	0.36	0.19	0.29	0.49	0.49	0.45	0.09
Brain	0.34	0.49	0.44	0.44	0.43	0.42	0.04
Heart	0.35	0.58	0.38	0.34	0.33	0.35	0.09

Table 3. Day to day variation of the mean percentage body burden per gram as measured by the standard error. Each value is the mean of all animals for all routes sacrificed on a given day

Organ or Tissue	1 day	2 days	7 days	S.E.
Kidney	5.36	5.36	5.58	0.34
Salivary glands	0.95	1.06	0.98	0.18
Testes	0.69	0.81	0.77	0.04
Muscle	0.73	0.76	0.67	0.08
Lymph nodes	0.72	0.70	0.48	0.14
Bone	0.66	0.67	0.65	0.02
G. I. tract	0.67	0.50	0.63	0.13
Heart	0.64	0.63	0.50	0.13
Spleen	0.58	0.51	0.58	0.21
Liver	0.49	0.53	0.52	0.04
Respiratory system	0.49	0.51	0.43	0.17
Hair	0.38	0.12	0.78	0.25
Skin	0.33	0.32	0.36	0.03
Brain	0.24	0.32	0.26	0.04

or no thallium indicates a rapid equilibration with tissues, which is also compatible with this premise. A material very soluble in body fluid would be expected to leave the site of injection very rapidly and there is some evidence that this happens. The blood of an animal injected intravenously contained no detectable thallium at approximately 20 min after injection, and the respiratory system of an animal injected intratracheally showed little thallium at 2–3 hr after injection.

Excretion

Daily fecal to urinary ratios are presented in Fig. 2 as calculated from 3-day weighted averages. The similarity between all routes is seen, although there are some minor differences. A slightly higher ratio is apparent following IP injection, and a lower ratio following SQ injection. These variations may well be due to slight differences in rate of loss from the injection site. The rate of decrease in excretion is greater in the urine than in the feces as was found by other investigators.[4,5] There are no data for the IV route after 7 days because collection of urine and feces was discontinued for a period of 1 week and owing to the low concentration of thallium injected by this route, the values after two weeks were insignificant.

MPC's for Tl^{204}

The fact that the body burdens (as percentage administered dose) from all routes fall into the same population at any time, and that they follow a single exponential function which extrapolates to 100 per cent at zero time, indicates a complete absorption from the

Table 4. Route to route variation of the mean percentage body burden per gram as measured by the standard error. Each value is the mean of all animals sacrificed on 1, 2 and 7 days for a given route of administration

Organ or tissue	IV	IP	IM	SQ	IT	O	S.E.
Kidney	5.20	5.47	6.09	5.71	5.42	4.71	0.24
Salivary glands	0.49	1.02	1.28	1.39	0.71	1.08	0.13
Testes	0.54	0.67	0.86	0.81	0.78	0.88	0.03
Muscle	0.62	0.71	0.76	0.71	0.73	0.79	0.06
Lymph nodes	0.47	0.85	0.58	0.70	0.52	0.68	0.10
Bone	0.58	0.63	0.70	0.66	0.67	0.74	0.02
G. I. tract	0.91	0.56	0.50	0.49	0.52	0.62	0.09
Heart	0.42	0.91	0.63	0.53	0.53	0.54	0.09
Spleen	0.35	0.28	0.53	0.60	1.01	0.56	0.15
Liver	0.47	0.53	0.48	0.52	0.53	0.52	0.03
Respiratory system	0.34	0.55	0.52	0.50	0.49	0.49	0.12
Hair	0.07	0.54	0.36	0.97	0.25	0.37	0.18
Skin	0.29	0.33	0.33	0.33	0.37	0.37	0.02
Brain	0.18	0.30	0.30	0.29	0.28	0.27	0.02

Table 5. Mean per cent body burden per organ and per gram wet weight with standard errors

Organ or tissue	per organ	S.E.	per gram	S.E.
Muscle	33.66	0.70	0.72	0.03
Bone	20.15	0.46	0.66	0.01
G. I. tract	13.33	0.80	0.60	0.05
Skin	9.59	0.40	0.34	0.01
Kidney	8.12	0.23	5.43	0.14
Liver	4.32	0.10	0.51	0.02
Testes	2.74	0.09	0.76	0.02
Respiratory system	0.95	0.07	0.48	0.07
Hair	0.76	0.13	0.43	0.10
Salivary glands	0.56	0.04	1.00	0.07
Brain	0.43	0.02	0.27	0.02
Spleen	0.38	0.05	0.56	0.08
Lymph nodes			0.63	0.06
Heart	0.39	0.04	0.59	0.05

gastrointestinal tract. This is also true for thallium deposited in the respiratory tract by IT injection. Because of this complete absorption from lung and gastrointestinal tract the lung model,[12] used for calculation of an MPC for air, is in error. However, it makes no difference in the final outcome whether lung-deposited thallium is assumed to be completely absorbed to the blood or whether two-thirds of it is presumed to enter the gastrointestinal tract and be absorbed completely from there.

The biological half-time of any organ has to be equal to the biological half-time for body burden when there is no day to day variation in relative organ content. Such is the case with thallium (with the possible exception of the hair) as seen in Tables 1 and 3. Kidney has been chosen as the critical organ for dosage calculations because it contains an average of 5.4 per cent of the body burden per gram which is a factor of 5.4 greater than that of the next highest organ (Table 5). Using these assumptions, the components of the equations for MPC for air and water[13] are: $f_1 = 1.00, f_2 = 0.054$, $T_b = 3.3$ days, $T_e = 3.3$ days and $f_{2'} = f_2$. The mass of the human kidney, 300 g, and E_{max}, 0.76 MeV, were used in the calculations for m and $\Sigma(bE)$, respectively.

With kidney as critical organ, the maximum permissible body burden (q) is equal to 18.7 μc, the maximum permissible concentration in air $(MPC)_a$ for a 40-hr week is 7.5×10^{-7} μc/cm^3, and the maximum permissible concentration in water $(MPC)_w$ for a 40-hr week is 5.2×10^{-3} μc/ml.

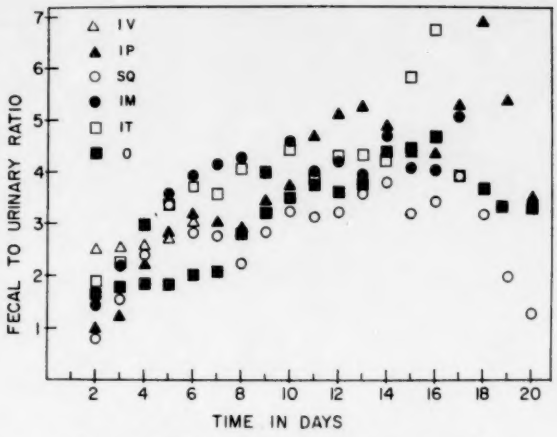

FIG. 2. Daily thallium excretion expressed as a ratio of fecal to urinary concentration.

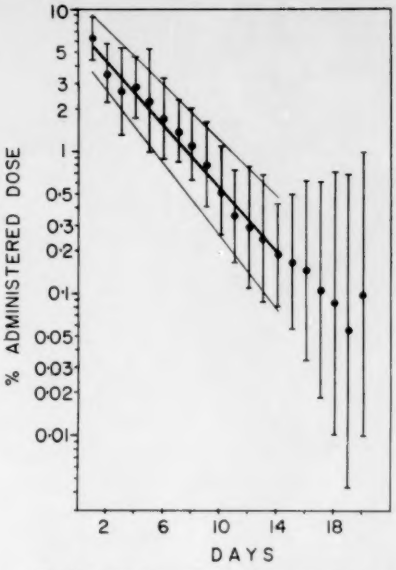

FIG. 3. Urinary excretion as percentage administered dose showing 75 per cent tolerance intervals.

Bio-assay

Urinary excretion as percentage administered dose per day is presented in Fig. 3. As with the tissue distribution data, the excretion rates from all routes appear to be from the same population. Seventy-five per cent tolerance intervals with 95 per cent confidence are also given[14] and least squares lines are drawn through the mean excretion values and through the upper and lower tolerance limits. The tolerance intervals may be used in calculating administered dose ("zero time" body burden) from urinary excretion values following intake of unknown amounts of thallium. On any given day the urinary excretion level of at least 75 per cent of a population having received thallium of unknown quantity at a known time will fall within the tolerance limits shown, with 95 per cent confidence. Using the lower limit, the highest probable dose received can be calculated, whereas, using the upper limit, one can calculate the lowest probable dose.

If one plots body burden for single instantaneous doses received each day, using the kinetics of retention ($T_b = 3.3$ days) shown in Fig. 1, an equilibrium is established at about 20 days. However, because of this short biological half-time, weekends should be considered when the data are used to simulate an industrial exposure schedule. When this is done for single daily exposures 5 days per week, an equilibrium condition is not reached until approximately 30 days. Because of the sizeable decrease in body burden over a weekend, there is build up through the working week such that the maximum body burden is reached on Friday of each week and this "equilibrium" body burden (after 30 days) is equivalent to 4.4 times the daily dose. Therefore, a suitable bio-assay method would be analysis of a Monday morning urine sample for thallium which corresponds to 2.67 days after exposure (Friday afternoon). As seen in Fig. 3 the urinary excretion rate at 2.67 days is 3.6 per cent of the administered dose (Friday afternoon body burden) per day.

If y = urine excreted per day = 1.5 l., z = amount of urine in Monday morning sample, x = amount of Tl in Monday morning sample, and BB = body burden on Friday afternoon

then xy/z = amount of Tl/day at 2.67 days = 3.6 per cent body burden, BB = $(x/z)(y/3.6\%)$ = 41.6 (x/z).

If the maximum permissible body burden calculated on the basis of continuous exposure 7 days a week (18.7 μc) were used as the maximum permissible Friday afternoon body burden with the exposure schedule just described, the Monday morning urine sample corresponding to this value would be 0.45 μc/l.

Using the tolerance limits shown for 2.67 days, the level of Tl/l. of urine at that time has an upper limit of 0.77 μc beyond which the body burden is very apt to be over the calculated MPC.

It is recognized that the calculations presented are based solely on data obtained from rats and may not be applicable to human exposure. However, in the absence of sufficient human data either to support or refute these calculations they may reasonably be assumed to approximate correct values.

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AN APPROACH TO THE QUESTION OF COMPUTING DOSES AND EFFECTS FROM FALL-OUT

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Abstract—Estimates of the possible number of injuries that might occur as a result of fall-out from nuclear weapons tests are frequently presented in scientific literature. In most cases the emphasis is put on the numerical result, which invites criticism, since the validity of certain assumptions, especially those regarding the biological effect of small doses cannot be proved.

The great number of uncertainties involved in such estimations leads many authors to feel that rough approximations are justified. However, a number of important parameters are thus often overlooked, and certain assumptions necessary to the results are accepted without being recognized.

This paper is an approach to the question of the parameters and assumptions that must be taken into account in such calculations; parameters and assumptions which would in fact influence the final considerations of the actual effects of radiation on a population. Provided that the physical fall-out mechanism is known, it is possible to calculate various organ doses that might be relevant to the biological effects. Calculations of the dose in the gonads and the bone marrow have been made, with special regard to the two extreme assumptions of biological dose effect relation, namely a linear relation without threshold, and a threshold relation. This survey of the parameters and assumptions leads to some interesting results; e.g. that there are indications that the *per capita* mean marrow dose in a population may be relatively little affected by the actual retention mechanism of Sr^{90} in bone.

INTRODUCTION

THE following approach to a general method of computing doses and effects from fall-out was first elaborated in connection with studies undertaken by the United Nations Scientific Committee on the Effects of Atomic Radiation.[1] A preliminary draft of this material was prepared and circulated in the course of the work of the Committee in 1958. At this stage, it was studied and worked over intensively by many individuals from a number of countries whose comments and criticisms greatly improved the document and materially assisted the author in preparing the present version of it. Thanks are due to all of these individuals, but especially to Dr. R. K. APPLEYARD, who initiated the work, and to Drs. R. BJÖRNERSTEDT, C. L. COMAR, D. H. COPP and A. KJELBERG, who very actively participated in the preparation of the original version. The International Business Machines kindly assisted in making some of the more involved numerical computations.

The purpose of the calculations in this paper is to assess the *possible* total incidence of deleterious biological effects of fall-out from nuclear test explosions under some specified assumptions, the validity of which cannot yet be verified. The calculations are presented in order to show that numerous assumptions have to be made and to illustrate the lack of knowledge at many important steps, rather than to provide a numerical result. The latter must therefore be regarded with great reserve although it is important to realize that the figures presented here inevitably follow from the assumptions made.

Where the biological effect on a population, expressed as a number of specified incidents of an injury, is the object of the computation, one must consider the time-lag caused by the long retention of radioactive material in the stratosphere. There is also a prolonged period in which uptake of radioactivity and subsequent radiation exposure occurs. Although a third

time-lag, namely that between the exposure and the incidence of the injury is common to all sources, most sources other than fall-out produce or initiate biological effects to a degree that can be measured by the extent of the radiation exposure at any time. In the case of fall-out, however, exposure at a certain time already implies subsequent exposure at other times. The total biological consequences of test explosions carried out up to a given date must therefore be evaluated by computing the future inevitable dose, as well as the present and past exposures.

Calculation schemes based upon each of the following two assumptions relating radiation dose with possible biological effect are presented in this paper :

(a) There is no threshold of the relevant dose, and the dose-effect relation is linear.

(b) There exists a threshold of the relevant dose.

It must be appreciated that these assumptions probably give much too simplified a picture of the mechanism and merely constitute two extreme alternatives. It is possible that the true dose-effect relation is far more complicated. However, even the treatment of relatively simple extreme assumptions may prove helpful for the understanding of the actual process. If the dose-effect relations will be found to be non-linear or in some cases dependent on age, the formulae and constants will have to be changed, but the general approach will still be valid or at least provide a helpful basis for a reappraisal of the situation.

Case of non-threshold, linear dose-effect relation

The "dose" relevant to the total genetic effect on the population following a certain pattern of nuclear test explosions is taken to be the infinite time integral of the gonad dose rate, assuming that all individuals will be exposed to the same dose-rate irrespective of age and sex.

The corresponding "dose" relevant to the estimate of the possible radiation-induced occurrence of leukemia, caused by the assumed time distribution of the environmental contamination, will be assumed to be the infinite time integral of the *per capita* mean marrow dose rate, taking into account the influence of age and life expectancy. As will be seen in the following, the same result can be derived by a more direct method of calculation, which has been preferred here.

The above doses, multiplied by the number of population and an appropriate constant, would give the total number of affected individuals in the population for all time during and subsequent to the exposure. All these calculations necessitate a number of assumptions which will be stated in detail in the following text.

No scientific prediction of the future practice of weapon tests is possible. However, as long as a linear dose-effect relation is assumed with no threshold, each year of testing must result in the same ultimate total number of injuries, if the rate and manner of injection of long-lived fission products into the stratosphere are the same. It is therefore appropriate to compare radiation doses and, where possible, the biological effects for one year of each practice that will, whether at the time or subsequently, give rise to exposure.

For the total effect it is irrelevant *when* the exposure occurs, and only the total relevant dose need be known. A year of practice should therefore be considered as related to a total *dose commitment* rather than to some actual exposure over a given period.

Case of threshold assumption

In the threshold case, on the other hand, the comparison of actual exposures is necessary to consider whether the sum of the contributions exceeds the threshold at any time. For that reason, estimated exposures over appropriate periods of time may be made. As it may also be of individual interest to know when the mutational damage will enter the population at the maximum rate, 30-year integrals of the gonad dose have been calculated.

Assumptions made for the purpose of calculations

The uncertainty and complexity of the calculations may be exemplified by the computation of the relevant bone marrow dose due to Sr^{90} from stratospheric fall-out, where the following steps are involved:

(1) Assumption on rate of injection of Sr^{90} into the stratosphere.

(2) Assumption of mechanism and numerical expression for depletion of the stratospheric reservoir.

(3) Calculation of the rate of Sr^{90} fall-out.

(4) Assumptions regarding the geographical distribution of fall-out and estimate of a population-weighted average.

(5) Calculation of the accumulated fall-out deposit.

(6) Assumption of future influence of weathering.

(7) Numerical assumptions concerning food chain transmission of Sr^{90} and calculation of the contamination of diet.

(8) Numerical assumptions concerning uptake and retention of Sr^{90}.

(9) Calculation of accumulated Sr^{90} concentration in bone as a function of calendar date and date of birth.

(10) Numerical assumption connecting Sr^{90} concentration in bone with marrow dose, including assumption on the relative location of Sr^{90} with relation to the location of active marrow. Assumptions on the size distribution of the marrow cavities must be made.

(11) Assumption regarding the relevant dose for estimates of a possible induction of leukemia.*

(12) Assumption on dose-effect relationship and possible weighting factors for age, marrow size (mass of active marrow) or other parameters of biological significance.

(13) Calculation of the relevant dose rate.

(14) Calculation of the relevant individual dose.

(15) Calculation of the relevant *per capita* dose (including appropriate weighting factors) due to a certain contamination of the environment.

(16) Integration of the *per capita* dose over all environmental contributions due to a certain injection.

* This again is on the assumption of a correlation between marrow exposure and the induction of leukemia.

THE FALL-OUT FUNCTIONS

Radioactive material that has once been injected into the stratosphere will fall out over the earth's surface according to a mechanism, which is not yet known. Regardless of this mechanism, the material balance is governed by the following two equations:*

$$\frac{d\bar{Q}(t)}{dt} = n - \lambda\bar{Q} - \bar{F}_r(t) \qquad (1)$$

$$\frac{d\bar{F}_d(t)}{dt} = \bar{F}_r(t) - \lambda\bar{F}_d(t) \qquad (2)$$

where $\bar{Q}(t)$ = stratospheric content of any long-lived radioactive isotope, expressed per unit area of the surface of the earth (mc/km² per year); n = rate of injection of the isotope into the stratosphere, expressed per unit area of the surface of the earth (mc/km² per year); λ = disintegration constant of the radioactive isotope; $\bar{F}_r(t)$ = world-wide average fall-out rate of the isotope per unit area (mc/km² per year); $\bar{F}_d(t)$ = world-wide average accumulated deposit of the isotope per unit area (mc/km² per year).

The equations (1) and (2) cannot be solved unless an assumption with regard to the relation between $\bar{Q}(r)$ and $\bar{F}_r(t)$ is made, and it is also necessary to make some assumption with regard to the function for the variation of n with time. The simplest assumption with regard to the first relation would be that the rate of depletion of radioactive material from the stratosphere at any time is proportioned to the quantity stored there, i.e.

$$\bar{F}_r(t) = k\bar{Q}(t) \qquad (3)$$

There is no meteorological justification for the above assumption but it can be used for rough estimates until the true mechanism is known. Equation (3) implies that there is a certain mean residence time $1/k$. Recent estimates indicate that this time might be shorter than 1 year even though a value of 10 years was earlier considered possible.[2–7] The value will depend upon where, geographically, the injection is made, and the shortest resident

* Equation (2) implies that no leaching or weathering occurs.

times seem to be related to injections near the poles. In the report of the UNSCEAR the value $k = 0.1$/year was used, primarily since it was assumed to yield results on the pessimistic side. A value of $k = 0.35$ corresponding to a mean residence time of about 3 years would, however, be more realistic, and it must be remembered that it is likely that a "mean residence time" as defined above does not even exist.

In the following, the functions $\bar{F}_r(t)$ and $\bar{Q}(t)$ will be used to denote the fall-out rate and stratospheric content of Sr^{90}. They can be used to describe approximately also the fall-out rate and stratospheric content of Cs^{137} because of the close agreement in decay periods and similarity with respect to gaseous precursors within the two radioactive chains in question; the only correction that is necessary is the multiplication by the ratio of fission yields.

Geographical factors

$\bar{F}_r(t)$, $\bar{F}_d(t)$ and n in the equations are not local values but averaged over the whole surface of the earth. Such geographical averages are not relevant for estimates of the biological effects of a specific population (i) or on the whole population of the earth. Although not used in these calculations, appropriate local values of $F_r^{(i)}(t)$ and $F_d^{(i)}(t)$ might be derived by help of a geographical factor $G_i(t)$, such that

$$F_r^{(i)}(t) = G_i(t)\,\bar{F}_r(t) \qquad (4)$$

$$F_d^{(i)}(t) = G_i(t)\,\bar{F}_d(t) \qquad (5)$$

The geographical factor $G_i(t)$ as a function of latitude is shown in Fig. 1, as estimated for the years 1956–1957. The shape of the latitude variation will depend on the injection site. Measurements show that radioactive material injected into the stratosphere close to the Equator will be distributed in both the Northern and Southern Hemispheres, with about twice as much going to the Northern Hemisphere as to the Southern Hemisphere if the injections are slightly north of the Equator. Radioactive material injected into the stratosphere near the North Pole seems to deposit almost entirely in the Northern Hemisphere.[7]

A value $\bar{G}(t)$ appropriate for the whole population of the earth can be derived by

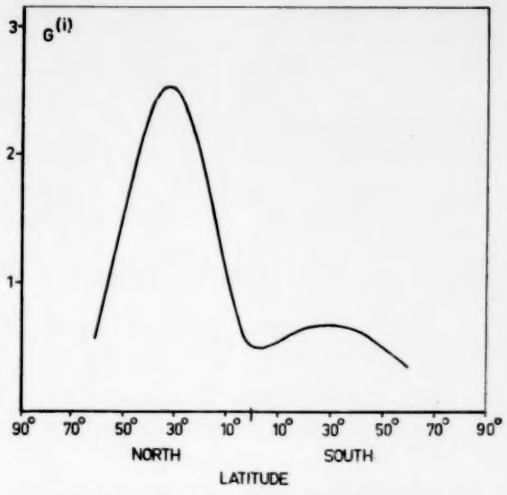

FIG. 1. Schematic presentation of the latitudinal variation of $G^{(i)}$ for the years 1956–1957 as assumed in the Report of the UNSCEAR.[2] Local meteorological factors have not been taken into account.

averaging through summation over all populations N_i. The value for mid-1958 has been calculated as

$$\bar{G}(1958) = \frac{\Sigma G_i(1958)\,N_i}{\Sigma N_i} \approx 2 \qquad (6)$$

It is possible that the local variation of $G_i(t)$ will change after the cessation of injection, and that $\bar{G}(t)$ may approach unity. However, as the geographical variation of the fall-out rate in the future cannot at present be predicted, it has been assumed, for the purpose of the following calculations, that at any time $\bar{G}(t) \approx 2$. The injection in the Northern Hemisphere in the fall of 1958 gave rise to a fall-out that seems to have had a comparatively high time constant of depletion and also increased the value of $G_i(t)$ in these regions more than at the Equator or in the Southern Hemisphere.

General expressions of the fall-out functions

In the case that the injection rate is constant, it is possible to integrate the equations (1) and (2), the general solution for the *population-weighted average* being

$$F_r(t) = \bar{G}(t)\,\bar{F}_r(0)e^{-\Lambda t} + \bar{G}(t)\,\frac{kn}{\Lambda}\,(1 - e^{-\Lambda t}) \quad (7)$$

$$F_d(t) = \bar{G}(t)\,\bar{F}_d(0)e^{-\lambda t} + \bar{G}(t)\,\frac{\bar{F}_r(0)}{k}\,(e^{-\lambda t} - e^{-\Lambda t}) + \bar{G}(t)\,\frac{n}{\lambda}\left(\frac{k}{\Lambda} + \frac{\lambda}{\Lambda}e^{-\Lambda t} - e^{-\lambda t}\right) \quad (8)$$

where $\Lambda = k + \lambda$.

Assumptions with regard to the continuation of nuclear test explosions

Any prediction of future fall-out rates must rely upon arbitrary assumptions with regard to the future pattern of testing. It is obvious that each year of testing with the same stratospheric injection will ultimately produce the same effect whenever the tests occur, as long as the dose-effect relation is linear with no threshold. It is therefore helpful to evaluate equilibrium effects on the assumption that the tests will continue at a certain rate, since the annual effect occurring in an equilibrium must be equal to the total effect produced by each year of testing. If the equilibrium effect is expressed per unit injection rate (i.e. the equilibrium effect calculated on any assumption is divided by the corresponding injection rate) the value should be fairly independent of the assumption of fall-out mechanism for long-lived substances ($k \gg \lambda$). On the basis of this value the total effect of any sequence of tests up to a given time can be calculated, provided the injection pattern is known.

First in the case of the threshold assumption (or when the burden to a population during a particular period of time, e.g. during the worst affected year, is the object of the study) the actual time distribution of dose must be known. In this case it is necessary to know $\bar{F}_r(t)$, $\bar{F}_d(t)$ and $G^{(i)}(t)$ at any time, and the result becomes very sensitive not only to the assumptions on k and $\int n\,dt$ but also to the assumption on $n(t)$.

For the linear dose-effect relation without threshold it can be shown that $F_r(\infty)$ and $F_d(\infty)$, normalized to unit injection rate, are the relevant parameters with regard to the total effect. From equations (7) and (8) they can be derived as

$$\frac{1}{n}F_r(\infty) = \frac{k}{\Lambda}\,\bar{G} \quad (9)$$

$$\frac{1}{n}F_d(\infty) = \frac{1}{\lambda}\cdot\frac{k}{\Lambda}\cdot\bar{G} \quad (10)$$

If $k \gg \lambda$, most of the injected material will fall out before the decay and the two expressions reduce to $\bar{G}$ and $(1/\lambda)\bar{G}$, respectively.

In the case of the threshold assumption, where information with regard to $F_r(t)$ and $F_d(t)$ is required also at finite values of t, assumptions must be made on the actual function $n(t)$, the most simple assumption, for illustrating purposes, being that $n(t) = \text{const} = n$ from $t = 0$ up to a certain time $t = T$, and $n(t) = 0$ for $t > T$.

For the numerical examples, values of n have been chosen that were assumed to be of the same magnitude as the annual injection from the test explosions already made. With knowledge of the number of "megatons" of fission energy related to stratospheric injection, and the absolute fission yield of Sr^{90} per megaton, it would be possible to calculate the total injection of Sr^{90} for each year up to date. n can also be calculated from known values of $\bar{F}_r(t)$, k and λ, provided that the assumption on the meteorological mechanism is correct. The latter estimate has been made on two different assumptions.

Assumption (a): The fall-out will remain constant and equal to $\bar{F}_r(0)$. In this case the condition defined by equation (3) will imply a constant stratospheric content (mainly due to large initial injections), maintained by a compensating constant injection rate.

Assumption (b): The injection rate will remain at a constant value equal to the mean value for the five years 1954–1958 inclusive.

The above two arbitrary assumptions, with regard to the future injection rate, yield the following expressions for n:

$$n_a = \left(1 + \frac{\lambda}{k}\right)\bar{F}_r(0) \quad (11)$$

$$n_b = \frac{\lambda}{1 - e^{-5\lambda}}\left(\bar{F}_d(1958) + \frac{\bar{F}_r(1958)}{k}\right) \quad (12)$$

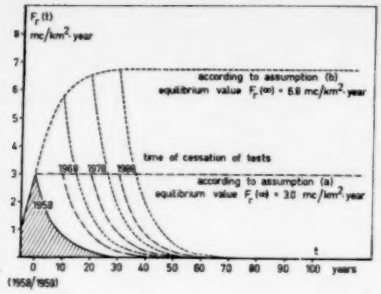

FIG. 2(a) Fall-out rate (population-weighted world average) following the assumptions made by the UNSCEAR (cf. page 345) with regard to injection rate and stratospheric retention.

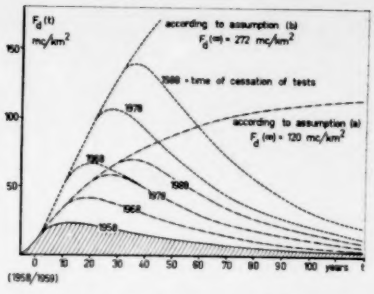

FIG. 2(b) Fall-out deposit (population-weighted world average) following the assumptions made by the UNSCEAR (cf. page 345) with regard to injection rate and stratospheric retention.

Particular expressions for specified intervals of time

The expressions given in the preceding section can be used for evaluation of the future fall-out under the particular assumptions made. For each assumed value of n, the functions $F_r(t)$ and $F_d(t)$ can be derived for a set of values of T. In these calculations it is necessary to consider separately the following three intervals of time:

(A) Period before $t = 0$ (i.e. the time at which the computation is made).

(B) Period between $t = 0$ and the time of cessation of injection.

(C) Period after time of cessation.

For $t < 0$ the best available approximation of the actual values of $\bar{F}_r(t)$ and $\bar{F}_d(t)$ should be used, as based on field observations. For $t > 0$, the following expressions are valid.

Period between $t = 0$ and $t = T$. For the period between $t = 0$ and the time of cessation of injection, the equations (7) and (8) can be rearranged to read:

$$F_r(t) = \bar{G}\frac{kn}{\Lambda} - \bar{G}\left(\frac{kn}{\Lambda} - \bar{F}_r(0)\right) e^{-\Lambda t} \quad (13)$$

$$F_d(t) = \bar{G}\frac{1}{\lambda}\frac{kn}{\Lambda} + \bar{G}\left(\frac{n}{\Lambda} - \frac{\bar{F}_r(0)}{k}\right) e^{-\Lambda t} - \bar{G}\left(\frac{n}{\lambda} - \frac{\bar{F}_r(0)}{k} - \bar{F}_d(0)\right) e^{-\lambda t} \quad (14)$$

Period after cessation of injection. If all injection is ceased at the time $t = T$, the fall-out functions will, for all subsequent time, be

$$F_r(t) = \bar{G}\bar{F}_r(T)\, e^{-\Lambda(t-T)} \quad (15)$$

$$F_d(t) = \bar{G}\left(\bar{F}_d(T) + \frac{1}{k}\bar{F}_r(T)\right) e^{-\lambda(t-T)} - \bar{G}\frac{1}{k}\bar{F}_r(T)\, e^{-\Lambda(t-T)} \quad (16)$$

Numerical expressions for $F_r(t)$ and $F_d(t)$ have been given in the original document, assuming the following values for the constants

$k = 0.1$/year
$\lambda = 0.025$/year (for both Sr^{90} and Cs^{137})
$\Lambda = 0.125$/year
$\bar{F}_r(0) = 1.5$ mc/km² per year
$\bar{F}_d(0) = 5$ mc/km²
$\bar{G} = 2$

With these assumptions, the values of n_a and n_b according to equations (11) and (12) will become 1.875 mc/km² per year and 4.25 mc/km² per year, respectively. It was assumed that $t = 0$ at the end of 1958.* The value $k =$

* The USSR test explosions in the Arctic region in October 1958 were not included in this calculation. The total fission energy of these tests has been estimated at at least 20 "megatons" whereas the total fission energy earlier released by UK, USA and USSR is said to have been about 70 "megatons".[7] If about 2/3 were injected into the stratosphere this would mean an average injection rate over the period 1954–1958 (five years) of the order of 10 "megatons" per year. This corresponds to a total activity of about 1 Mc Sr^{90}/year,[10] which gives $n = 2$ mc/km² per year. If the USSR tests were included, the value would be $n \approx 2.5$ mc/km² per year. Such estimates, however, are uncertain, since the assumed Sr^{90} fission yield depends upon assumptions with regard to the type of weapon tested and can vary by a factor of 2.[23]

0.1/year is lower than values which have been suggested more recently. A value of $k = 0.35$ would yield $n_a = 1.6$ and $n_b = 2.0$, which is still a reasonable figure with regard to the available information on the fission energy which has hitherto been released, and not very different from the values assumed for the first calculations. According to equation (9) the equilibrium value of $F_r(t)$ will be $\approx n$ for any values of $k \gg \lambda$. Although the shape of the fall-out functions for $t > T$ will be quite different with different values of k, this does not significantly influence the integrated effect of isotopes with long half-life such as Sr^{90} and Cs^{137}, provided that the value of $\bar{G}$ is not affected (e.g. by rapid fall-out over a small area or over sparsely populated regions).

The numerical expressions for $F_r(t)$ and $F_d(t)$, with the assumed values of the constants listed above, resulted in the curves shown in Fig. 2.

GONAD EXPOSURE

Dose-effect relationship

In this paper the genetic effect of exposure from fall-out will be estimated on the assumption that the dose-effect relation is linear without threshold, and that all gonad dose contributions up to the time of reproduction are additive.

The number of individuals affected subsequent to a certain gonad exposure will be estimated as the product of the population involved (which will be assumed to be constant), the relevant dose and a dose-effect constant. The relevant dose is the population average of the (by child-expectancy) weighted gonad dose. A dose increment δD will result in δN individuals affected in the future, computed as

$$\delta N = K_g P \delta D \tag{17}$$

The genetic dose-effect constant K_g can be derived from estimates of the representative doubling dose $\bar{D}_2$ and the current fraction f of adversely affected births. In genetic equilibrium the doubling dose, if received by every individual member from conception to the mean age of child-bearing, will give rise to fB affected births per year, where B is the birth-rate.

For the following calculations a model population (of constant size P) could be considered, where the age of breeding is 30 and the age of death is 70 years for all members. As B would then be approximated by $P/70$, the relation between the rate of introducing affected individuals and the rate of population exposure would be

$$\frac{dN}{dt} = \frac{3}{7} fP \frac{1}{\bar{D}_2} \frac{dD}{dt} \tag{18}$$

In the general case, however, the ratio w/W should be introduced instead of 3/7, where w is the *future* child expectancy of the average individual in the population and W is the *total* number of children expected by this individual including those already born.

Hence, not only in genetic equilibrium but as a result of any dose increment δD, the total subsequent number of affected individuals will be

$$\delta N = \frac{w}{W} fP \frac{1}{\bar{D}_2} \delta D \tag{19}$$

and, therefore, the genetic dose-effect constant is

$$K_g = \frac{w}{W} f \frac{1}{\bar{D}_2} \text{ rem}^{-1} \tag{20}$$

In the case of stratospheric fall-out, the gonad exposure may be extended over many decades even if the injection is ceased now. Each year of injection will give rise to an extended exposure, i.e. will imply a certain *dose commitment*. For calculation of the total number of genetically affected individuals over subsequent years (N_g) the appropriate dose related to any pattern of injection is the infinite time integral of the population average of the weighted gonad dose rates. If this dose is denoted D_∞,

$$N_g = K_g P D_\infty \tag{21}$$

For the purpose of this paper it will be assumed that the gonad dose rate at any time is the same for all individuals regardless of age and sex. No weighting for child expectancy is then necessary and the population average is equal to the individual dose.

Gonad exposure from stratospheric fall-out

Stratospheric, tropospheric and local fall-out contribute to the gonad exposure. As the tropospheric contribution each year is

correlated to the rate of tropospheric injection, it can be considered as a constant annual contribution over the time during which this injection is maintained constant. This will be discussed in a later section. The stratospheric fall-out that contributes to the gonad exposure is assumed to be mainly Cs^{137}. The fall-out functions derived in the preceding section can therefore be used as the basis for the dose computation if allowance is made for the difference in fission yield between Sr^{90} and Cs^{137} in the case of fission of U^{238}.

The gonad dose from Cs^{137} will result from both external and internal exposure.

External contribution. The following expression can be derived for the gonad dose rate when the source is a deposit $F_d^j(t)$ of a certain isotope (j) in a layer under an infinite plane.

$$\frac{dD_j}{dt} = kB_jS_jF_d^j(t) \text{ mrem/year} \qquad (22)$$

where k_j = dose rate from primary radiation (mrad/year per mc/km^2)
B_j = ratio total/primary radiation dose rate (build-up factor)
S_j = shielding factor
$F_d^j(t)$ = deposit of isotope j (mc/km^2)

In the general case, the primary dose rate at a distance h cm above the ground and assuming the active material uniformly distributed within an infinite layer of depth d cm, covered by an inactive layer of thickness s cm, e.g. snow, can be derived as (8)

$$k_j = \frac{723}{d}\sum_i E_{ji}n_{ji}\frac{(\mu_i - \sigma_{si})_{\text{air}}}{\mu_{ig}}\{x_i\mathbf{Ei}(-x_i) - (x_i + y_i)\mathbf{Ei}(-x_i - y_i) + e^{-x_i}(1 - e^{-y_i})\} \text{ mrad/year per mc/km}^2 \qquad (23)$$

where E_{ji} = quantum energy (MeV)
n_{ji} = number of quanta of energy E_i per primary disintegration
$(\mu_i - \sigma_{si})_{\text{air}}$ = true absorption coefficient of air for quanta of energy E_i (cm^{-1})
μ_{si} = total absorption coefficient of non-active covering layer for quanta of energy E_i (cm^{-1})
μ_{gi} = total absorption coefficient of ground for quanta of energy E_i (cm^{-1})
$\mu_{i\text{air}}$ = total absorption coefficient of air for quanta of energy E_i (cm^{-1})

$$x_i = (h - s)\mu_{i\text{air}} + s\mu_{si}$$
$$y_i = d\mu_{gi}$$
$$\mathbf{Ei}(-x) = -\int_x^\infty \frac{e^{-t}}{t}\,dt$$

In the case of Cs^{137}, equation (23) reduces to

$$k_{\text{Cs}} = \frac{0.189}{d\rho_g}\{x\mathbf{Ei}(-x) - (x + y)\,\mathbf{Ei}(-x - y) + e^{-x}(1 - e^{-y})\} \text{ mrad/year per mc/km}^2 \qquad (24)$$

where ρ_g is the volume weight of the ground material.

If the deposit is only on the surface, equation (23) becomes

$$k_{oj} = -723\sum_i E_{ji}n_{ji}(\mu_i - \sigma_{si})_{\text{air}}\,\mathbf{Ei}(-x) \text{ mrad/year per mc/km}^2 \qquad (25)$$

and

$$k_{o\text{Cs}} = -0.0146\,\mathbf{Ei}(-x) \text{ mrad/year per mc/km}^2 \qquad (26)$$

Equations (24) and (26) are presented graphically in Fig. 3. As a result of weathering, the value of k may decrease, and if part of the radioactive material becomes completely unavailable because of this effect, an appropriate term should be included in equation (2) to take account of this loss.

In addition to the dose rate from the primary radiation there is a considerable contribution from scattered radiation. The resulting build up factor may be about 2,[8] but is difficult to derive theoretically.

Also the shielding factor is difficult to estimate for the practical case where there is a combined effect of body shielding and structural shielding. In practice also the topography of the surface or layer that carries the radioactive material will affect the result. The final result will therefore not be very accurate, and this is especially the case if the external exposure from the deposit is integrated over a long period after the cessation of nuclear test explosions. In the report of the UNSCEAR a factor of 0.1 was used for the combined effect of shielding and weathering.

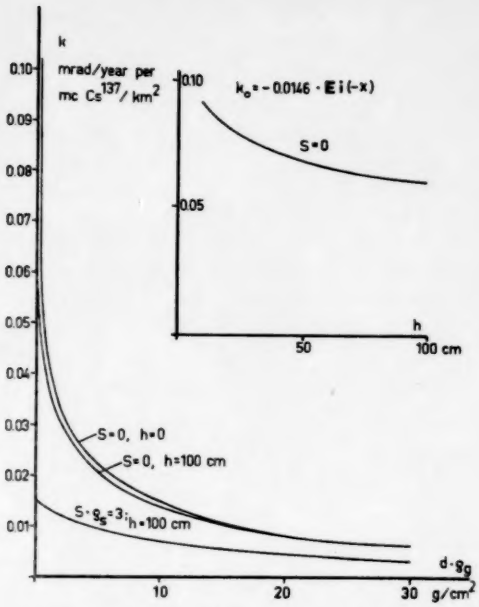

FIG. 3. Primary dose rate k in soft tissue (body shielding, structural shielding and build-up neglected) for various heights (h) over the ground, when the source is uniformly distributed in an infinitely wide layer of varying thickness (d) covered by an inactive layer (thickness s). k_0 denotes the primary dose rate when $d = 0$.

Internal contribution. In the case of exposure from internal sources the dose to the gonads will arise mainly from radioactive material incorporated in the soft tissues. At a point surrounded by a radioactive material uniformly distributed over a volume sufficient to produce saturation, the dose rate (neglecting differences in mass stopping power) will be

$$\frac{dD_j}{dt} = 18.7\bar{E}_j c_j \quad \text{mrad/year} \tag{27}$$

where $\bar{E}_j$ = mean energy emitted (and absorbed) per primary disintegration (MeV)
c_j = concentration of isotope j ($\mu\mu$c/g)

The mean energy emission $\bar{E}_j$ is composed of contributions from both β- and γ-radiation. The β component is the mean β-energy $\bar{E}_\beta$ (since there is no neutrino absorption) plus the converted fraction of the γ-energy. The γ component is the total γ energy, corrected for conversion. For Cs^{137} the corresponding energies are 0.23 and 0.54 MeV, respectively, i.e.

$$\left(\frac{dD}{dt}\right)_{Cs}^{\beta\infty} = 4.3c_{Cs} \quad \text{mrad/year} \tag{28}$$

$$\left(\frac{dD}{dt}\right)_{Cs}^{\gamma\infty} = 10.1c_{Cs} \quad \text{mrad/year} \tag{29}$$

For the β-radiation, saturation conditions exist in most cases. This is, however, not so for the γ-radiation, where there will be no energy equilibrium between the γ-emitting mass element and its environment. The γ dose at a point P within a volume of unit density containing a uniformly distributed radioactive material must be calculated by integration over all volume elements at different distances r

$$\left(\frac{dD}{dt}\right)^{\gamma P} = kc\int_v \frac{e^{-\mu_{\text{eff}}\, r}}{r^2}\, dV = kcg_P \tag{30}$$

where the dose rate is obtained in mrad/year if

$k = 0.0876R$ (mrad/year per $\mu\mu$c/g)
R = dose rate at 1 m from 1 c (rad/hr)
c = concentration of radioactive material ($\mu\mu$c/g)

The geometrical factor g_P has been calculated for various points in the human body; the values on the "central axis" ranging from 60 to 180 with an average of about 120.[9] If the value 120 ± 60 is applied with regard to the gonad dose from Cs^{137} ($R = 0.29$ rad/hr), the γ contribution from internal Cs^{137} is estimated as

$$\left(\frac{dD}{dt}\right)_{Cs}^{\gamma} = (3.0 \pm 1.5)c_{Cs} \text{ mrem/year} \tag{31}$$

and the total gonad dose rate from internal Cs^{137}

$$\left(\frac{dD}{dt}\right)_{Cs} = (7.3 + 1.5)c_{Cs} \text{ mrem/year*} \tag{32}$$

The relation between the Cs^{137} concentration in the human body and the fall-out functions is not yet known in detail. Cs^{137} is taken up and excreted fairly rapidly by the body, but is poorly

* This is lower than the value based upon $\Sigma E(\text{RBE})n = 0.59$ that is usually applied to the whole body[13] and which would yield $dD/dt = 11c_{Cs}$.

taken up from soil by plants. Therefore, both the contamination of food sources and the concentration in the body have been assumed to be proportional to the fall-out rate, i.e.

$$c_{Cs} = \text{const. } F_r(t) \tag{33}$$

Experimental data on body burden of Cs^{137} ($c_{Cs} \approx 0.1$ $\mu\mu$c/g at a time when $F_r(t)$ was about 3 mc/km^2) indicate that the constant would be about 0.03–0.04 $\mu\mu$c/g per mc Cs^{137}/km^2 per year.[10–12]

Formula for total dose-rate in the gonads. From the above assumptions it follows that the gonad dose rate can be written as

$$\frac{dD}{dt} = g_i F_r(t) + g_e F_d(t) \tag{34}$$

where the constant g_i can be derived from equations (22) and (24), and g_e can be derived from equations (32) and (33).

If the functions $F_r(t)$ and $F_d(t)$ representing population-weighted averages are used, the dose rate will be applicable for computation of the relevant value of D_∞ for the whole population of the earth.

Equation (34) has been evaluated numerically in the original UN document,[1] assuming $g_i =$ 0.3 mrem/year per mc/km^2 per year and $g_e =$ 0.01 mrem/year per mc/km^2 and the fall-out functions as shown in Fig. 2. The result is presented in Fig. 4. Each year of injection at a certain rate implies a dose commitment that can be derived from the equilibrium values of the fall-out functions, calculated for the hypothetical case that this injection rate is maintained for infinite time.

$$D_\infty{}^1 = g_i F_r(\infty) + g_e F_d(\infty) \tag{35}$$

30-year integrals of gonad dose rate. As it may be of individual interest to know when the possible mutational damage will enter the population at the maximum rate, 30-year integrals D_{30} of the gonad dose have been calculated. The variation of D_{30} with time of birth is shown in Fig. 5, based on the dose rate curve in Fig. 4.

Contribution from tropospheric fall-out

Although the mechanism for depletion of the stratospheric reservoir of radioactive fission products is not known, the future fall-out has been estimated in the preceding paragraphs, assuming an exponential retention. As it is likely that the effective retention time is much smaller than the mean radioactive life of the most important γ-emitter, Cs^{137}, this assumption will not critically affect the estimate of the infinite time integral of the gonad dose rate. As the maximum 30-year gonad dose will be received by individuals born during the decade 1950–1960, if tests are not continued, this estimate is also fairly reliable whatever the actual mechanism of depletion may be, provided that $\bar{G}$ is known.

In the case of tropospheric fall-out, on the other hand, the assumption on the fall-out mechanism will critically affect the estimate, since the type of isotopes deposited will depend on the time constants involved. As the computation of genetic effects on the linear dose-effect non-threshold assumption is based upon a population-weighted average for the whole of the earth, certain geographical variations must be assumed. As the tropospheric fall-out deposit in equilibrium is proportional to the tropospheric injection rate, estimates of future deposit must be based upon assumptions on this rate in addition to the assumption on the stratospheric injection rate.

It may be assumed that recently observed levels of tropospheric fall-out deposit approximately represent an equilibrium of the most significant isotopes, characteristic for the last few years' pattern of testing. A continuation of this pattern of testing will therefore be expected to maintain the tropospheric fall-out deposit as well as rate. The geographical distribution will depend on the location of the test sites and on meteorological conditions.

The main source of uncertainty in the computation of a population-weighted average of the tropospheric fall-out is that not even the geographical distribution during the previous years of testing is sufficiently well known. However, local high values of deposit, which must be considered in all cases where a threshold assumption may be valid, may have no relevance in the estimate of *genetic* effects to the population as a whole.

External exposure. The external contribution

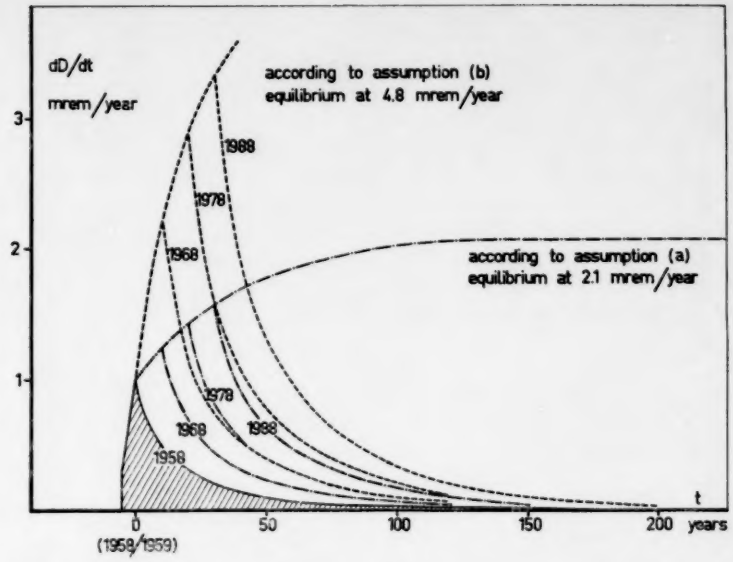

FIG. 4. Gonad dose rate from external and internal caesium-137 (population-weighted world average) calculated on the assumptions on injection rate and stratospheric retention made in the report of the UNSCEAR.

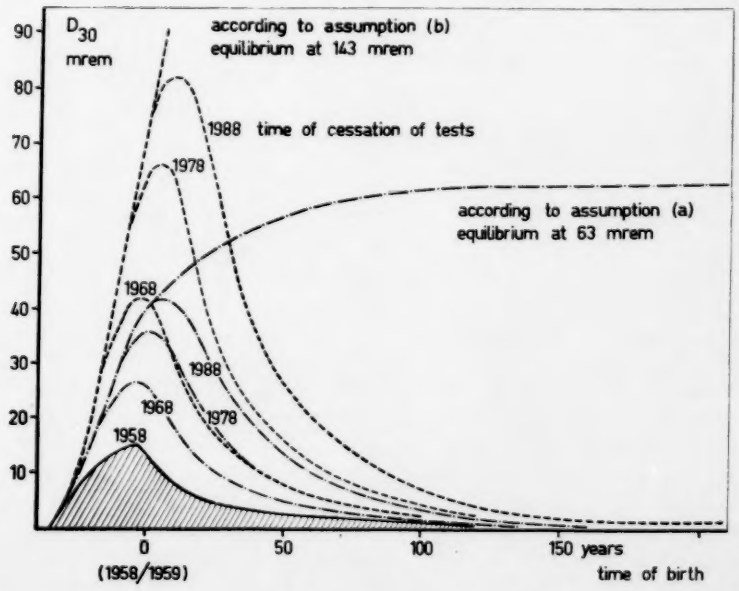

FIG. 5. 30-year gonad dose as derived from Fig. 4, given as a function of time of birth (population-weighted world average).

to the gonad dose from tropospheric fall-out can be calculated by equation (22) provided that the composition of the active deposit is known. In the U.N. report[2], the equilibrium at the actual injection rate for the years 1955–1957 is estimated at about 50–200 mc/km^2. Assuming an average γ energy of 0.5 MeV and a factor 0.1 for the combined effect of shielding and weathering, the annual gonad dose would be of the order of 0.25–1 mrem. It must be emphasized that this estimate is much more uncertain than the estimate of the present dose distributions from stratospheric fall-out.

Internal exposure. In the case of short-lived radioactive isotopes, the gonad dose from inhaled material may not be insignificant in relation to other contributions. Formulae and constants for calculation of dose from inhaled radioactive material have been prepared by the International Commission on Radiological Protection.[13] Assuming an air concentration of fission products at ground level of 10^{-15} c/l. the annual gonad dose from inhaled material would be of the order of some tenths of a mrem.

Few data are available to permit an estimate of the gonad exposure from short-lived fission products taken up with food and water. As a gamma exposure comparable with that from Cs^{137} in the body tissues would be detectable in whole-body γ-spectrography, the lack of evidence for such concentration of short-lived γ-emitters might be taken to indicate that the total internal exposure gives less than 1 mrem/year.

Total gonad dose from tropospheric fall-out. Because of the scant information, the estimate of the total annual gonad dose from tropospheric fall-out is very uncertain. It will be assumed here that this contribution will be 0.5 mrem as an average for the world's population, at a tropospheric injection rate such as for the years before 1958. As the tropospheric fall-out will disappear when the injection has ceased, the resulting dose rate is maintained only during the injection period.

An annual dose of 0.5 mrem during the period of injection implies a dose commitment of 0.5 mrem/year of injection to the troposphere at the assumed rate. In general, the tropospheric dose commitment per year of test can be related to the stratospheric injection rate by the formula

$$D^1_{\text{trop}} = k_{\text{trop}}\, n_{\text{trop}} = K_{\text{trop}}\, n \qquad (36)$$

provided that the ratio n_{trop}/n is known.

Total gonad dose from all fall-out

It is convenient to use the dose commitment per year of test at unit injection rate as a basis for computation of effects on the linear dose-effect relation assumption. For the combined effect of stratospheric and tropospheric fall-out, the value will be

$$\frac{1}{n} D_\infty{}^1 = g_i \frac{1}{n} F_r(\infty) + g_e \frac{1}{n} F_d(\infty) + K_{\text{trop}} \qquad (37)$$

Since $F_r(\infty)$ and $F_d(\infty)$ are proportional to n, the first two terms will not depend on the value assumed for n. The third term will be

$$K_{\text{trop}} = \frac{D^1_{\text{trop}}}{n_a} \qquad (38)$$

since n_a is the best estimate of the injection rate during the period for which the numerical value of D^1_{trop} has been derived. Equation (38) is an approximation of (36) only if the ratio n_{trop}/n is assumed to be the same for future tests.

With the values and assumptions used in the previous calculations, the numerical value of the dose commitment per unit injection rate will be

$$\frac{1}{n} D_\infty{}^1 = 0.48 + 0.64 + 0.27 \approx 1.4 \text{ mrems}$$

Contribution from C^{14}. The contribution to the dose commitment from the increase of C^{14} has not been included in the above calculations. An estimate in the original U.N. document[1] indicated annual gonad doses from this source of the order of 0.005–0.01 mrem. In spite of this low figure, the long mean life of this isotope—8000 years—would imply a dose commitment of some 40–80 mrems, which is higher than any other contribution. However, it is obvious that a dose most of which will be delivered very far in the future must be weighted properly before it is compared with doses delivered within relatively short periods, comparable with the human life-time.

BONE MARROW EXPOSURE

Dose effect relationships

The exposure of the bone marrow will be evaluated in the light of the possible induction of leukemia. Two assumptions will be treated in parallel because they give extreme results rather than because they are more likely than other assumptions. For both it will be postulated that the effect does not depend upon rate of exposure or fractionation of the dose. Any recovery has thus been disregarded. The assumptions are: (A) There exists a threshold of the relevant dose, and (B) there is no threshold, and the dose-effect relation is linear.

It should be noticed that the general approach is, in principle, applicable to any dose-effect relationship.

Threshold case. Where the existence of a threshold is assumed, it is appropriate to examine whether the highest exposure gives rise to a dose exceeding the threshold value. If this is not the case, the margin of safety can be estimated, i.e. it is possible to say by how much the mean exposure level can be increased before the threshold is exceeded. For this purpose, even if the average dose in a population is calculated, the spread around this average also needs to be assessed in some way.

The relevant dose (in the case of the threshold assumption) will be taken to be the 70-year mean marrow dose, averaged over the whole of the active marrow. This dose may not be relevant when only part of the marrow is exposed, but as the long-term marrow exposure due to fission products incorporated over a long period in the bone is fairly uniform, the 70-year mean marrow dose will be used here for the purpose of computation.

The true relevant dose may be the maximum dose to active marrow cells, wherever it occurs. However, no exposures can occur in bone marrow longer than over the life-time, and the 70-year dose is therefore adopted here in the absence of proof that a shorter period is appropriate.

If the total energy of a Sr^{90} body burden equivalent to 1 $\mu\mu$c Sr^{90}/g of calcium is assumed to be dissipated in 7000 g of bone, the average dose rate would be 2.64 mrems/year. It will be assumed here that some of the energy is delivered to the 1500 g of red bone marrow but the energy delivered to the yellow marrow or to tissues outside the skeleton will be disregarded. The mean values of the dose rates to bone marrow and spongious bone have been taken to be 0.6 and 0.9 mrem/year, respectively.[14,15] Assuming that spongious bone constitutes 10–12.5 per cent of the total bone mass, this implies dose rates of 2.7 and 2.5 mrems/year to compact bone and total bone, respectively (cf. Table 1).

In considering the maximum dose rates for cases A and B, the geometrical distribution of the uniform labeling is the only factor which has been taken into account.[14] For bone marrow the maximum dose rate will be obtained within Haversian systems inside compact bone. The difference between the values 2.7 and 3.0 in Table 1 represents a difference in stopping power. The maximum values due to uneven distribution are based only on differences in mineralization between regions with linear

Table 1. *Mean and maximum dose rates in* mrem/year *for chronic poisoning with* 1 $\mu\mu$c Sr^{90}/g *of calcium*[14]

	A. Uniform labeling		B. Uneven distribution factor = 2	
	Maximum	Mean	Maximum	Mean
Bone marrow	3.0 (1.8)	0.6	5.9 (3.6)	0.6
Spongious bone	1.7	0.9	3.4	0.9
Compact bone	2.7	2.7	5.4	2.7
Compact and spongious bone	2.7	2.5	5.4	2.5

Figures in brackets give the maximum bone marrow dose inside spongious bone.

dimensions of the same order as the Y^{90} β-range. The influence of microscopic changes in the mineralization has not been allowed for, yet it is believed that this is relatively unimportant, most probably less than a factor of 2. The ratio of maximum to mean marrow dose will be about 10, which includes the factors 2, for uneven distribution of the Sr^{90}, and 5, for the increased dose-rate in enclosed cavities.

Knowledge of the threshold level would imply evidence in support of the threshold theory. No such information exists, and all that can be said from the available data is that a possible threshold may lie anywhere between 0 and 400 rems.[16]

Case of non-threshold, linear dose-effect relation. On the alternative extreme assumption that no threshold exists and that there is a linear dose-effect relation, the number of persons who will develop leukemia after a certain marrow exposure can be estimated as the product of the population involved (which will be assumed to be constant), the appropriate dose and a dose-effect constant. The appropriate dose is the *per capita* mean marrow dose, where "mean marrow dose" denotes the average dose over the whole mass of active marrow. An increment δD of the *per capita* mean marrow dose will be assumed to result in a number δN of leukemia cases in the future, computed as:

$$\delta N = K_l \, P \delta D \qquad (39)$$

where P is the population and K_l the dose-effect constant for inducing of leukemia.

An estimate of the dose-effect constant for induction of leukemia can be made from the present data on number of occurrences following a certain exposure.[2] It will be assumed that a single exposure of 1 rem to the entire red marrow will result in 1.5 annual cases per million exposed for a period of 15 years following the exposure.* The total number of cases is therefore 23 per million exposed, and

$$K_l = 23/\text{rem per million}$$

This factor can be applied to all individuals having a life expectancy of more than 15 years at the time of exposure. In a model population where the age of death is 70 years, it is hence valid for 80 per cent of the population. In the following it will be taken to apply to the population as a whole.

Marrow exposure from stratospheric fall-out

In the case of stratospheric fall-out, the most important source of marrow exposure is Sr^{90} incorporated in the bone. There is also a marrow exposure from Cs^{137}, both external and internal. The marrow exposure from Cs^{137} varies with time in the same way as the gonad exposure from this source, and is approximately of the same magnitude. It is therefore described by the curves in Fig. 4. As it amounts to only about 5 per cent of the marrow exposure from Sr^{90}, it will be disregarded in the following estimate, and the marrow dose from Sr^{90} will be taken as the only significant contribution from stratospheric fall-out.

The Sr^{90} marrow exposure differs from the Cs^{137} gonad exposure not only in magnitude but also in the distribution over time. The gonad exposure is a function of the fall-out rate and deposit at the time of exposure, since Cs^{137} is fairly rapidly excreted. Because of the long retention of Sr^{90}, however, the marrow will be exposed over a long period of time following the uptake, and the relation between the fall-out functions and the marrow exposure at a certain time is relatively complicated.

Total bone burden of strontium-90. The relationship between the Sr^{90} body burden of the population and that of the dietary may be approximated by assuming that all of the calcium in the body will at any time have a strontium burden reflecting that of the dietary.[2]

* If the risk of developing leukemia is not limited to the first 15 years following the exposure, but maintained at constant value throughout life, the increment dN_k will be different for each age-group k of the population exposed to a dose dD. The corresponding dose-effect factor would be $1.5(70 - \tau_k)$/rems per million where τ_k denotes the age at the time of exposure. However, the available data seem to indicate a decrease of the annual rate of incidence after 10 years[16,17] and this possibility will not be considered here. As the mean age in a model population is 35 years, the average dose-effect factor would be $35 \times 1.5 = 52$/rems per million, which differs from the assumed factor only by a factor of 2.3.

This is obviously an overestimate, but is probably not in serious error for younger individuals because of the time pattern of Sr^{90} contamination. A more rigorous and general evaluation is possible from mathematical consideration of the rates of calcium and strontium accretion in relation to the projected time-patterns of Sr^{90}/Ca ratios. Such treatment provides two advantages: (a) more realistic values for the Sr^{90} burden are forthcoming and (b) general relationships in regard to age, date of birth, and pattern of Sr^{90}/Ca ratios with time that permit estimations of mean marrow doses.

Sr^{90} is taken up by the body through food-chains. The change in the ratio Sr^{90}/Ca during each step from soil to bone is often described by the help of discrimination factors, but for the purpose of these calculations the rate of Sr^{90} uptake will be assumed to follow the empirical expression;[2]

$$c(t) = a_r F_r(t) + a_d F_d(t) \qquad (40)$$

where $c(t) =$ ratio Sr^{90}/Ca in the minerals entering bone; a_r and $a_d =$ constants with different values for various food-chains.

Equation (40) makes it possible to determine the quantity q of Sr^{90} that is incorporated in the bone at the same time as a certain mass (α) of calcium:

$$q(t) = c(t)\alpha \qquad (41)$$

In the following, q and α will denote the rate of incorporation of Sr^{90} and calcium, respectively. The *net* rate of increase will depend on the rate of turnover of earlier incorporated material.

By using an empirical formula of the type (40), provided that it may be extrapolated over a hundred years, the bone burden of Sr^{90} can be computed by help of $c(t)$ and a number of assumptions on the calcium and strontium metabolism. It will be shown that the final result, in form of the infinite-time integral of the dose-rate, or the maximum 70-year dose, does not depend critically on the assumptions about the metabolism of bone.

The rate at which dietary Ca and Sr^{90} enter the skeleton depends on several processes the most important of which are formation of new bone mineral and resorption of existing bone mineral. For the system under consideration the processes of surface exchange of ions between bone and blood can be neglected. The uptake of calcium can then be approximated by equations based on the difference between rates of accretion and resorption.

Some evidence is available as to the way in which the human body content of calcium increases with age.[18] This evidence may be adequately approximated for the purposes of these calculations by assuming that the body calcium increases linearly with age from 6 months after conception up to age of 20 years. The errors introduced by replacing the sigmoid growth curve by a straight line with a discontinuity at age 20, are of the less importance since the main concern is with the dose integral to the age of 70.

Bone-salt metabolism has been studied in detail and models of varying complexity have been proposed.[19–21] The simple model described below is in keeping with the principles reflected in the reference material. With the approximation of the body calcium increase, the net uptake of calcium is governed by the equations:

($\tau < 20$ years)

$$\frac{dB(\tau)}{d\tau} = \alpha - k_0(\tau)B(\tau) = \text{const.} = \beta \qquad (42)$$

where

$$B(\tau) = \beta\tau,$$

and $$\alpha = \beta + k_0(\tau)B(\tau) = \beta(1 + k_0\tau) \qquad (42a)$$

($\tau > 20$ years)

$$\frac{dB(\tau)}{d\tau} = \alpha - k_0(\tau)B(\tau) = 0 \qquad (43)$$

where $B(\tau) = \text{const.} = B_a$, and $\alpha = k_0(\tau)B_a$ (43a)

The symbols are: $\alpha =$ rate of total calcium accretion, $k_0(\tau) =$ fraction of calcium turned over per unit time at age τ, $\beta =$ constant net increase of calcium per unit time under age 20, $B(\tau) =$ mass of bone calcium at age τ, and $B_a =$ mass of bone calcium in an adult.

No satisfactory evidence is available as to how the fractional calcium turnover rates vary with age in man. The changes in turnover rates could possibly be represented by power functions,[22] exponential functions or series of exponential functions. However, there are no data that permit mathematical treatment. It is

necessary then to determine how different values of the turnover rate will affect the results obtained.

In the general expression

$$\frac{dB}{d\tau} = \alpha - k_0(\tau)B(\tau) \qquad (44)$$

it is useful to consider two extreme values for k_0:

(I) If $k_0 = \infty$, an infinite amount of calcium is taken up (and turned over) per unit time. This implies that the bone is continuously in complete equilibrium with its environment, and the apparent concentration of Sr^{90} in bone, $s(t,\tau)$, is at any time

$$s(t, \tau) = c(t) \qquad (45)$$

(II) If $k_0 = 0$, calcium is acquired only during the period of growth, at the rate β, and then retained during the life-time.

As any retention curve can be approximated by a series of exponential functions which represent various fractional turnover constants, the result obtained with these expressions may be expected to fall somewhere between the limits set by the extreme cases (I) and (II). As these extremes will be shown to give quite similar results, more accurate assumptions do not seem to be called for.

When assumptions for the calcium metabolism have been made, the bone burden of Sr^{90} can be calculated from the equation

$$\frac{dQ(\tau)}{d\tau} = q - k_1 Q(\tau) \qquad (46)$$

where q = uptake of Sr^{90} per unit time $= c(t)\alpha$, k_1 = fraction of Sr^{90} turned over per unit time, $k_1 = k_{Sr} + \lambda$ (where k_{Sr} is the biological turnover factor and λ is the radioactive decay constant) and $Q(\tau)$ = total amount of Sr^{90} in bone.

There is reasonably good evidence that the turnover rates of calcium and strontium are essentially equal. The influence of various assumptions on the strontium retention will be discussed in the following sections. The use of a constant k_1 in equation (46) implies that the retention of strontium in bone is an exponential function of time. The following calculations will be performed on the basis of this assumption, and the possible error introduced hereby will be estimated.

The apparent concentration of Sr^{90} in bone i.e. the ratio $s(t, \tau) = Q(t, \tau)/B(\tau)$, can now be computed by integration of equation (46), assuming that k_0 is also constant.

($\tau < 20$ years)

$$s(t, \tau) = e^{-k_1\tau}\frac{1}{\tau}\int_0^\tau c(t)\ (1 + k_0\tau)\ e^{k_1\tau}\, d\tau \qquad (47)$$

($\tau > 20$ years)

$$s(t, \tau) = e^{-k_1\tau}\left\{\frac{1}{20}\int_0^{20} c(t)(1 + k_0\tau)\, e^{k_1\tau}\, d\tau + k_0\int_{20}^{\tau} c(t)\ e^{k_1\tau}\, d\tau\right\} \qquad (48)$$

Empirical formulas for $c(t)$. To use equations (47) and (48) it is necessary to know $c(t)$. As mentioned above, empirical formulas may be derived from available data of the type (40). In practice it will be assumed that the fall-out rate dependent term can be disregarded in *long term extrapolations*. Hence the following equation will be used:

$$c(t) = aF_a(t) \qquad (49)$$

In the report of the U.N. Scientific Committee[2], data and constants have been developed that relate the accumulated fall-out deposit of Sr^{90} to the Sr^{90}/Ca ratio found in milk (PERRY, N.Y. and the United Kingdom) and rice (Japan). These values may be used in the computations of typical radiation doses, assuming that these two foods constitute the entire source of dietary calcium and Sr^{90}. The constant a can be derived from the Sr^{90}/Ca ratio in the two diets and the discrimination factor milk-bone to give the Sr^{90}/Ca ratio in the new material incorporated in bone.*

* This constant gives the Sr^{90} ratio in the newly formed bone. In the case of rapid excretion during a short period after the uptake (power function retention), it is assumed that Sr^{90} and Ca are excreted at the same rate. If this is the case, the ratio of the fractions that are retained for longer periods will remain the same, and equations (41), (42a) and (43a) will still give the amount of Sr^{90} that is taken up to be incorporated in bone over long periods of time. If this is not the case, $c(t)$ should denote the Sr^{90}/Ca ratio of the more firmly incorporated material. In the latter case, the constant a will have lower values if Sr^{90} is excreted more rapidly than calcium.

(1) For a milk diet:

$$a_M = 0.15\ \mu\mu\text{c Sr}^{90}\text{/g Ca per mc/km}^2$$

(2) For a rice diet:

$$a_R = 0.9\ \mu\mu\text{c Sr}^{90}\text{/g Ca per mc/km}^2$$

Marrow dose rate. The mean marrow dose rate will be proportional to the apparent concentration of Sr^{90}, $s(t, \tau)$.

$$\frac{dD(t, \tau)}{dt} = \gamma\, s(t, \tau) \tag{50}$$

The proportionality factor γ may be taken to be 0.6 mrem/year per strontium unit, whereas the dose-rate in the bone should be taken to be 2.5 mrems/year per strontium unit.

The uncertainty involved in the estimate of the best value of the dose constant γ is due mainly to lack of information on the location of the bone strontium with respect to the active marrow, the difficulty of calculating the actual dose distribution from the information of the strontium distribution, and to uncertainty with regard to the size distribution of the marrow cavities.

As the dose rate is a function of date of birth as well as of calendar date, it cannot be represented by a single curve for each assumption on injection practice. In the following sections, methods will be given for computation of the infinite time integral of the *per capita* mean marrow dose-rate which can be used for calculating the dose commitment for each practice: the maximum 70-year integrals may also be computed, so as to indicate the margin of safety in the case of the threshold assumption.

Infinite time integral. The dose commitment for a certain injection practice can be calculated as the infinite time integral of the *per capita* mean marrow dose-rate resulting from the practice studied. However, in the following, the dose commitment has been assessed by help of a different approach, considering the effect of an infinitesimal uptake of Sr^{90} at a certain time.

The Sr^{90} taken up into bone during an infinitesimal period dt, at time t, can be expressed by

$$dQ = \alpha\, c(t)\, dt \tag{51}$$

An invidual of age u at the time of uptake will at a later age τ have retained

$$dQ(u, \tau) = dQ(u, u)\ e^{-k_1(\tau - u)} \tag{52}$$

By integrating the dose rate caused by this retained Sr^{90} over the balance of life, one obtains the life-time dose increment to the marrow due to the individual uptake during the period dt at age u and time t:

$$dD(u, t) = F_m(u)\gamma\, c(t)dt \tag{53}$$

The dose increment factor $F_m(u)$ for individuals with an expected life span of m years will be shown to be:

Under age 20 years

$$F_m(u) = \frac{\alpha}{k_1 B_a}\,(1 - e^{-k_1(m-u)}) \tag{54}$$

Over age 20 years

$$F_m(u) = \frac{k_0}{k_1}\,(1 - e^{-k_1(m-u)}) \tag{55}$$

In the general case $(\alpha/B_a) = k_0(B(u)/B_a) + (\beta/B_a)$ and with the simplified linear model of bone growth $\alpha/B_a = (1/20)(1 + k_0 u)$, for $u <$ 20 years.

The value $d\bar{D}$ of the life time dose increment averaged over the whole population subject to the uptake during the period dt at time t can be calculated as

$$d\bar{D}(t) = \frac{1}{m}\int_0^m dD\ du = \gamma c(t)\ dt \bar{F}_m \tag{56}$$

where

$$\bar{F}_m = \frac{1}{m}\int_0^m F_m(u)\ du \tag{57}$$

With the average dose increment factor $\bar{F}_m$ known for a population where the mean life is m years, the dose commitment for an injection practice causing the contamination $c(t)$ can be calculated as

$$D_\infty = \bar{F}_m \gamma \int_{-\infty}^{+\infty} c(t)\ dt \tag{58}$$

The dose increment factor $F_m(u)$ is given in Fig. 6, assuming the accretion of skeletal calcium reported by MITCHELL *et al.*[18] The biological interpretation of the factor is discussed in the text to the figure. The average values $\bar{F}_{70}$, for

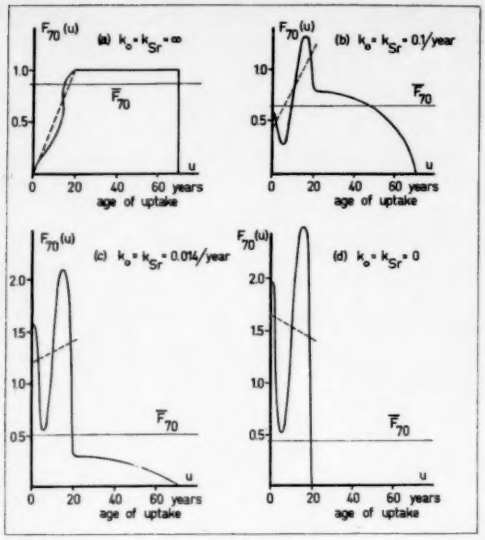

FIG. 6. The dose increment factor $F_{70}(u)$ for an expected life span of 70 years, as a function of age of uptake. (a) Extreme case of instantaneous equilibrium between bone and environmental Sr^{90}, with infinite turnover constant. Note the high dose this would give to the adult. (b) Biological half life for Sr and Ca assumed to be 7 years ($k_{Sr} = 0.1$/year). (c) Biological half life for Sr and Ca assumed to be 50 years ($k_{Sr} = 0.014$/year). Note the increasing importance of uptake and retention in childhood, and the reduced uptake in the adult. (d) Essentially no Sr and Ca turn-over. Uptake only during bone growth in childhood.

a model population where every member lives to age 70 years, are indicated in the diagrams. It will be shown that $\bar{F}_m$ is not critically dependent on either the rate of calcium and strontium turnover or the mean age of the population. The simple linear model for calcium accretion under age 20 years would give results indicated by the broken lines on Fig. 6.

The formulas (54) and (55) have been derived on a specific assumption about the significance of the exposure of marrow in children. On the non-threshold, linear dose-effect assumption, the mean marrow doses must be weighted for mass of marrow. The following two extreme cases have been considered, namely: (1) No weighting for the smaller amount of marrow in children, and (2) weighting of the child dose in proportion to the mass of bone. As there is more marrow in proportion to bone in a child than in an adult, the case (2) will give an overcorrection, and the true result can be expected to lie between the two cases (1) and (2). The influence of marrow weighting is illustrated by Fig. 7.

In the case that the marrow is weighted in proportion to the mass of bone, the formulas (54) and (55) can be derived from (53) where $dD(u, t)$ is the integral of the dose-rate caused by $dQ(u, \tau)$ over the balance of life:

$$dD(u, t) = \int_u^m \frac{d}{d\tau}(dD)\, d\tau \qquad (59)$$

and

$$\frac{d}{d\tau}(dD) = \frac{\gamma}{B_a}\, dQ\,(u, \tau) \qquad (60)$$

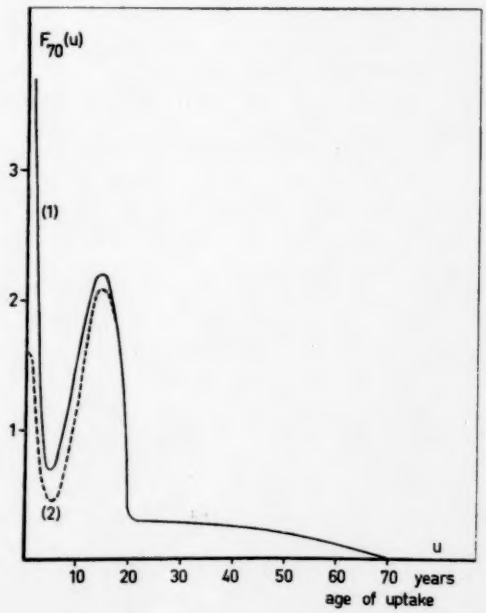

FIG. 7. The influence of marrow weighting on the dose increment factor $F_{70}(u)$, assuming a biological half life of 50 years for strontium and calcium. (1) No weighting for the smaller amount of marrow in children. (2) Weighting in proportion to the mass of bone in the child.

The integration gives

$u < 20$ years:

$$dD(u, t) = \gamma c(t)\, dt \frac{\alpha}{k_1 B_a} (1 - e^{-k_1(m-u)}) \quad (61)$$

$u > 20$ years:

$$dD(u, t) = \gamma c(t)\, dt \cdot \frac{k_0}{k_1} (1 - e^{-k_1(m-u)}) \quad (62)$$

In the case that no marrow weighting is made, the formula (60) must be replaced by the following expression for $u < 20$ years

$$\frac{d}{d\tau}(dD) = \frac{\gamma}{B(\tau)} dQ(u, \tau) \quad (63)$$

which with the simple linear model of bone growth leads to

$u < 20$ years

$$dD(u, t) = \gamma c(t)\, dt(1 + k_0 u) \times \left\{ e^{k_1 u} [-\mathbf{Ei}(-k_1 u) + \mathbf{Ei}(-20k_1)] + \frac{1}{20k_1} (e^{-k_1(20-u)} - e^{-k_1(m-u)}) \right\} \quad (64)$$

where $-\mathbf{Ei}$ denotes the exponential integral. Accordingly the dose increment factor will be

$$F_m(u) = (1 + k_0 u) \times \left\{ e^{k_1 u} [-\mathbf{Ei}(-k_1 u) + \mathbf{Ei}(-20k_1)] + \frac{1}{20k_1} (e^{-k_1(20-u)} - e^{-k_1(m-u)}) \right\} \quad (65)$$

The influence of the marrow weighting is shown in Fig. 7. All other diagrams have been derived from the formulas (54) and (55) and not from (65), i.e. they illustrate the marrow-weighted case.

Fig. 8 shows the dose increment factor $F_m(u)$ for the special case that $k_0 = k_{Sr} = 0.014$ and various values of the expected life (m).

Fig. 9 shows how $F_m(u)$ will be affected in the case when the turnover of strontium is more rapid than that of calcium.

Fig. 10 summarizes the results by showing how the average dose increment factor $\bar{F}_m$ varies with the turnover rate of calcium and the expected life in a population. The variation is apparently very limited, ranging from 0.5 to 0.8 for likely values of k and m.

As the $\bar{F}_m$ is not critically dependent on the assumptions on bone metabolism, the retention of calcium will be approximated, for the purpose of these calculations, by a single exponential function of time, with $k_0 = 0.014$, which corresponds to a half-life of 50 years. This is the value that has been suggested by the ICRP. If Sr^{90} is assumed to have the same biological half-life as calcium, the value of k_1 will be 0.04. For the following calculations $\bar{F}_m$ will be taken to be 0.5. If no correction for marrow mass is

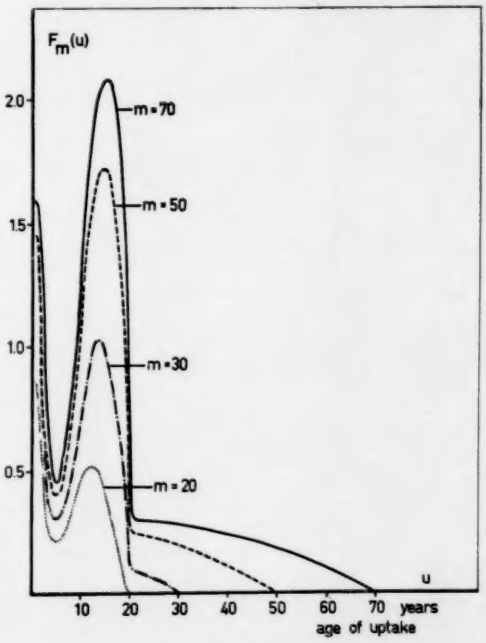

FIG. 8. The variation of the dose increment factor $F_m(u)$ with the expected life span (m), assuming a biologic half life of 50 years for strontium and calcium.

Note: With reduced expected life span, a large proportion of the population will be in the younger age group with high uptake and retention of Sr^{90}, but the time for irradiation of the marrow is correspondingly shortened.

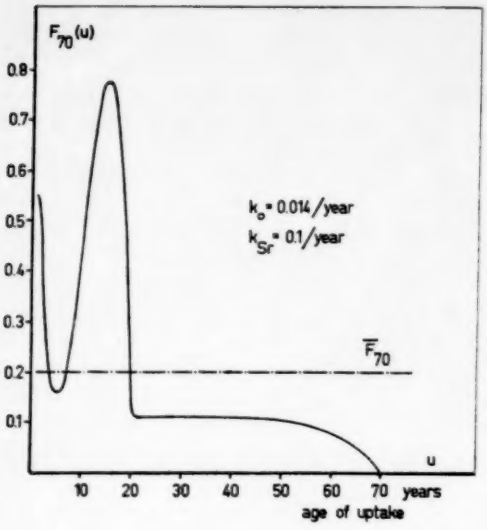

FIG. 9. The dose increment factor $F_{70}(u)$ as a function of age of uptake of Sr^{90}, assuming a biological half life of 50 years for calcium and 7 years for strontium.

Note: This represents an extreme case based on the assumption that strontium is turned over in the skeleton at a much faster rate than calcium. Animal experiments and data from the radium poisoning cases in man suggest that both turnover rates are extremely slow in the adult.

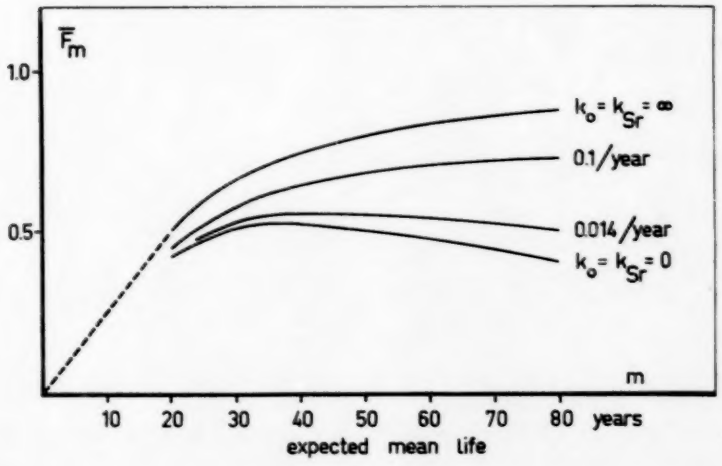

FIG. 10. The population average of the dose increment factor for various turnover constants as a function of the expected mean life.

Note: For life spans between 30 and 50 years and for values of $k_{Sr} = k_0$ in bone ranging from 0 to ∞, the range of the mean population dose increment factor $\bar{F}_m$ is only 0.5–0.8. The reason for this remarkably narrow range is the compensating effects of changes implied in the various assumptions which have been made. For example, a more rapid turnover will reduce the retention of strontium taken up by a child, but will increase the uptake and exposure of the adult. With a shorter mean life span for the population, a larger proportion will be in the younger age group with a high uptake of strontium, but the total life-time exposure is reduced.

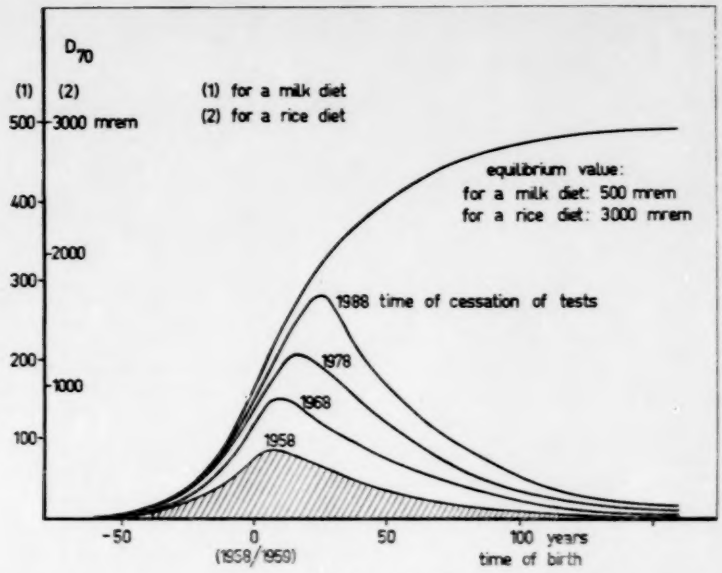

FIG. 11. 70-year *per capita* mean marrow dose (population-weighted world average) as a function of time of birth, if tests continue on assumption (a) of the UNSCEAR (cf. page 345).

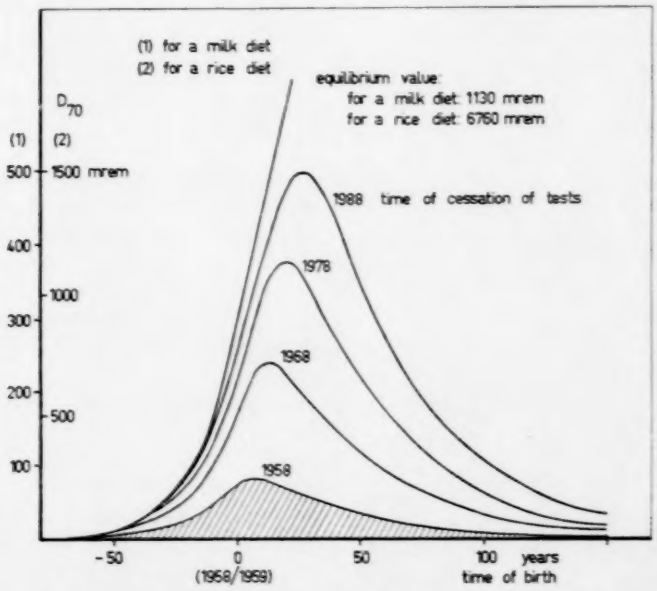

FIG. 12. 70 year *per capita* mean marrow dose (population-weighted world average) as a function of time of birth, if tests continue on assumption (b) of the UNSCEAR (cf. page 345).

made, the factor will be 0.65 for $m = 70$ years in the case illustrated in Fig. 7.

With $\bar{F}_m = 0.5$ and $\gamma = 0.6$ equations (58) and (49) give:

(1) For a milk diet

$$D_\infty = 0.045 \int_{-\infty}^{+\infty} F_d(t)\, dt \quad \text{(mrems per mc/km}^2\text{)} \tag{66}$$

(2) For a rice diet

$$D_\infty = 0.27 \int_{-\infty}^{+\infty} F_d(t)\, dt \quad \text{(mrems per mc/km}^2\text{)} \tag{67}$$

The dose commitment per year of injection must be the same whatever time t the injection occurs. It can therefore be calculated from equilibrium considerations, using equation (10) as

$$D_\infty{}^1 = \bar{F}_m \gamma a F_d(\infty) \tag{68}$$

which yields $19.2an$ mrems per year of injection at the rate n mc Sr^{90}/km^2 per year. The values for the constant a may be taken from page 357.

70-year integrals of the mean marrow dose-rate

The life-time mean marrow dose as a function of the time of birth (t_b) will be calculated as

$$D_{70}(t_b) = \int_0^{70} \frac{dD(t,\tau)}{d\tau}\, d\tau = \int_0^{70} \gamma s(t,\tau)\, d\tau \tag{69}$$

using the expressions (47) and (48) for $s(t,\tau)$. This is a numerically complicated calculation, which has been performed for various values of injection rate and time of cessation of injection. The result is shown graphically in Figs. 11 and 12.

Contribution from tropospheric fall-out

Fig. 13 gives an example of the variation of the dose increment factor $F_m(u)$ according to equations (54) and (55) for an uptake $\alpha c(t)\, dt$ of Sr^{89}, which is one of the most important isotopes to consider in the estimate of marrow exposure from tropospheric fall-out. With $\bar{F}_m = 5 \times 10^{-3}$ the dose commitment for a practice resulting in the contamination $c(t)$ will be

$$D_\infty = 5 \times 10^{-3} \gamma \int_{-\infty}^{+\infty} c(t)\, dt \tag{70}$$

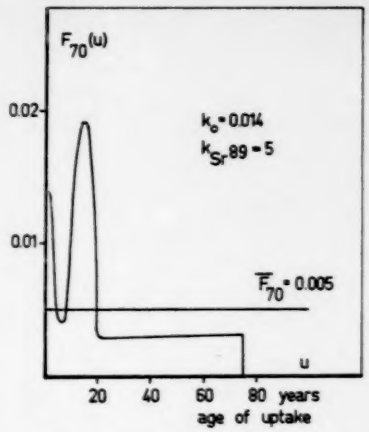

FIG. 13. The dose increment factor $F_{70}(u)$ in the case of Sr^{89} with a physical half life of 51 days ($k_{Sr^{89}} = 5$/year).

In the case of Sr^{89} the dose constant γ is taken to be 1 mrem/year per $\mu\mu$c Sr^{89}/g Ca. It will be assumed that $c(t)$ is 0.5 times the concentration of Sr^{89} in milk, for which Fig. 14 shows some reported data. The integral of $c(t)$ over 1 year can be estimated as about 20 $\mu\mu$c Sr^{89}/g Ca per year. Accordingly the dose commitment for 1 year of injection is

$$D_\infty{}^1 = 5 \times 10^{-3} \times 1 \times 20 = 0.1 \text{ mrem}$$

The formula (47) may be used for an estimate of the apparent concentration of Sr^{89} in a child. A simplified model of the seasonal variation of Sr^{89} in milk has been assumed (on the basis of the Canadian data[21] shown in Fig. 14) for calculation of the contamination of the bone of infants born at a time so that the calcification starts at the beginning of the peak of environmental contamination. For these infants, the mean marrow dose during the first year would be about 10 mrems,* and the 70-year mean marrow dose some 20 mrems if the tropospheric injection is continued. For children born at

* In the case of the *threshold* assumption, where the maximum marrow dose may be relevant, it is important to note that the maximum marrow dose for acute poisoning with radiostrontium may exceed the mean marrow dose by a factor of 25–50 compared with the factor 10 for chronic poisoning.[14]

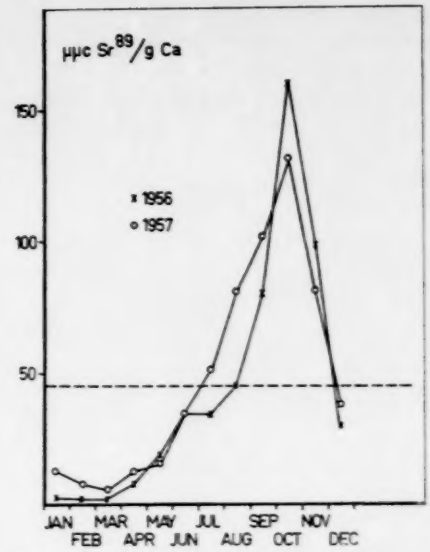

FIG. 14. Example on seasonal variation of the levels of Sr^{89} in milk (Canadian data).

other times of the year, however, the dose would be smaller, and the *per capita* mean marrow dose in the whole population, including adults, would be of the order of 0.1 mrem per year.* The calculated mean marrow dose rate for the infants is shown in Fig. 15.

POSSIBLE BIOLOGICAL CONSEQUENCES OF NUCLEAR WEAPON TESTS

Using the dose-effect constants and the calculated dose commitments per year of injection, it is possible to compute—on the assumptions presented above—the number of seriously affected persons in the world's population. These numbers cannot be said to represent the *expected* result, nor even the *likely* one, as no level of likelihood can be associated with the assumptions made. However, they are numbers that cannot be ruled out on the basis of present knowledge and they follow from the assumptions made in this text.

The *total* consequences in the form of the number of cases over *all* years subsequent to *one* year of injection of fission products to the stratosphere were, for the case of the assumption of a non-threshold, linear dose-effect relation, calculated in the original document from the

* See footnote, p. 362.

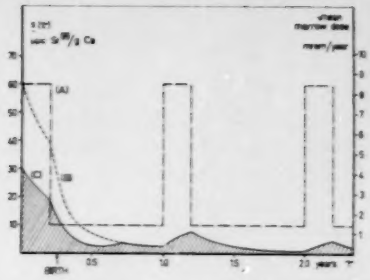

FIG. 15. Apparent concentration of Sr^{89} in bone of children for whom the calcification starts at the beginning of the seasonal contamination peak. (A) Assumed seasonal variation of Sr^{89} concentration in new material incorporated in bone, milk being the main source. (B) Apparent concentration $s(\tau)$ of Sr^{89} if milk is the main source from the start of calcification. (C) Same, corrected for placental discrimination and lower concentration in mother's milk.

following formulas summarizing the preceding computations.

For the number of major genetic effects

$$\frac{1}{n} N_g^1 = K_g P D_\infty^1 = K_g P \times \left\{ g_i \frac{1}{n} F_r(\infty) + g_e \frac{1}{n} F_d(\infty) + K_{\text{trop}} \right\} \quad (71)$$

where according to equation (38) $K_{\text{trop}} = D_{\text{trop}}^1 / n_a$, and the estimate of $F_r(\infty)$ and $F_d(\infty)$ depends upon the meteorological mechanism that is assumed. If $k \gg \lambda$, most of the injected Cs^{137} will fall out before the decay and equation (71) will become

$$\frac{1}{n} N_g^1 = K_g P \left\{ g_i \bar{G} + g_e \frac{1}{\lambda} \bar{G} + D_{\text{trop}}^{1,\text{gen}} / n_a \right\} \quad (72)$$

where n = world average injection rate (mc Cs^{137}/km^2 per year), N_g^1 = number of major genetic effects for all future years following one year of test at the injection rate n, $K_g = \frac{w}{W} f \frac{1}{D_2}$ /rem according to equation (20), P = the total population of the earth, g_i and g_e = constants in equation (34), λ = disintegration constant of Cs^{137}, $\bar{G}$ = population-weighted average of the geographical factor, equation (6).

Of the three terms within the brackets, the tropospheric contribution is the most uncertain. With the numerical values of the constants given in the text, the ratio of the terms would be 6 : 8 : 3. It may, however, be that the Cs^{137} contribution has been overestimated, and, especially when the maximum individual dose or dose-rate is of concern, it is important to realize that there is a much greater geographical variation of the tropospheric than of the stratospheric fall-out.

For the possible number of leukemia cases

$$\frac{1}{n} N_l^1 = K_l P D_\infty^1 = K_l P \left\{ \bar{F}_m \gamma \frac{1}{n} \times [a_r F_r(t) + a_d F_d(t)] + \text{trop. contr.} \right\} \quad (73)$$

where again, if $k \gg \lambda$ and the rate-depending term is neglected

$$\frac{1}{n} N_l^1 = K_l P \left\{ \bar{F}_m \gamma a \bar{G} \frac{1}{\lambda} + D_{\text{trop}}^{1,\,\text{m rrow}} / n_a \right\} \quad (74)$$

where N_l^1 = number of leukemia cases over all future years following one year of test at the injection rate n, K_l = dose-effect constant as defined by equation (39), P = the total population of the earth, $\bar{F}_m \approx 0.5$ as defined by equations (54), (55) and (57), γ = dose constant, see Table 1 and equation (50), a = food-chain constant, see equations (40) and (49).

In the above expression the tropospheric contribution may be neglected without introducing a substantial error in the calculation of the effect on the linear dose-effect non-threshold assumption. With the threshold assumption, however, the maximum dose to the marrow may be relevant in each individual case. The tropospheric contribution would then become significant both because of the local geographical concentration of tropospheric fall-out and because of the non-uniform distribution of bone-seeking radioisotopes in the skeleton.

The number of cases under the assumption of a threshold in the dose-effect relation can only be estimated from the number of individuals that would have a dose exceeding the threshold dose because of the total exposure produced by *all sources* of radiation. The number of cases of injury would be much less than the number of persons exceeding the threshold. The 70-year marrow doses, as shown in Figs. 11 and 12, may not be relevant in the threshold case but may have to be multiplied by a non-uniform distribution factor that is of the order of 10 for chronic and 25–50 for acute Sr^{90} poisoning. The population-weighted averages do not apply either, and the local values of the geographic factor G_i must replace $\bar{G}$.

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SOME PHYSICAL AND PHYSIOLOGICAL FACTORS CONTROLLING THE FATE OF INHALED SUBSTANCES—I*†

DEPOSITION

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Abstract—This paper discusses various physical and physiological factors in terms of their effect on aerosol stability and especially as they are related to the deposition of dusts in the respiratory tract of man. Although a unifying formulation of these factors is not available, nor attempted herein, efforts are made to indicate the nature of the more important factors and the circumstances in which they might be most significant.

I. INTRODUCTION

THE purpose of this paper is to bring together in one place a reasonably comprehensive discussion of some of the important physical and physiological factors which determine dust deposition.

Dust deposition can be defined as the active process wherein dust is removed from the air while it is being respired. The term implies the extent to which dust is removed in terms of the same measure of dustiness as used to characterize the inspired air. Some fraction of the dust deposited is not in reality on the surface membrane of the respiratory tract, but in suspension within the lung air. In the overall course of dust deposition the amount of dust so involved is relatively small and short-lived and is particularly insignificant when deposition is measured as a steady-state condition.

There are a variety of experimental approaches to the measurement of dust deposition. The common requirement in all methods is the ability to measure some parameter(s) of the airborne dust before and after it is breathed. The change that is usually measured is the change in the dust concentration. This results in a statement of percentage mass deposition. It is also possible to express deposition in terms of the number of particles or the number of particles in particular size regions.

To make dust deposition quantitative so that toxicity or retention studies can be pursued, it is necessary to measure, in addition to the aforementioned parameters, the amount of dusty air respired. For example, knowing the percentage mass deposition and the amount of air respired and its dust concentration (concentration in inspired air), the product of these three values will yield the amount of dust deposited in the respiratory tract. Special techniques are then employed if it is necessary to delineate the pattern of deposition, i.e. to know how much is deposited in the nasal-pharynx, how much is deposited in the lung parenchyma, etc. In many experimental situations, this can be approached by careful histologic and analytic efforts following sacrifice of the animal.

Certain radioisotopes which can be analyzed by external probes are now being widely employed to obviate the requirement for animal

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sacrifice and thereby, both the original sites of deposition and the course of the dust redistribution can be followed in the same subject.

The clearance or redistribution of dust from the respiratory tract bears a reciprocal relationship to dust retention. A discussion of some of the possible factors and mechanisms influencing retention will be given in Part II of this paper.*

II. PHYSICAL FORCES

Dust deposition in the respiratory tract can be thought of as a process occurring within a small compartment of air with the same physical forces in control that are involved in the dust deposition from the environmental air outside of the body. The forces of interest are of three general types; gravitational, inertial and Brownian motion.

A. *Gravitational settling*

A 20 cm dia. lead ball dropped from an airplane will be constantly accelerated toward the earth according to Newton's Law of Gravitation. A particle of lead of 20 μ dia. dropped into a chamber of tranquil air similarly falls by gravitation, but it is not continuously accelerated; it quickly reaches its terminal velocity and an acceleration of zero.

The dynamics of small particles (0.5–50 μ) falling through a fluid medium are controlled by the viscous forces of the medium. Stokes' Law states that the viscous resistance of a medium is equal to the product of the viscosity of the medium and the velocity and size of the particle.[1] The terminal velocity of fall is reached when the gravitational force and the viscous resistance force are equal.

For spheres in air, the gravitational force, F, is equal to $(\pi/6)d^3(\sigma - \rho)g$ and the air resistance or buoyant force, R, is equal to $3\pi\eta d\ V$ where d = dia. of particle, σ = density of particle, g = acceleration of gravity, η = viscosity of air, ρ = density of air, V = velocity of particle and $\pi/6$ and 3π are shape factors for spheres.

Setting these two equations equal to each other and solving for the terminal velocity of a particle yields the following equation:

$$V_s = \frac{(\sigma - \rho)gd^2}{18\eta}$$

The mobility of a particle, Z, is defined, under Stokes' Law conditions as equal to $1/3\pi\eta d$; therefore $V_s = ZF$. Stokes' Law conditions are those in which the ratio of the inertial to viscous forces is very small (<0.05).

As the particle diameter approaches the mean free path of air molecules, λ, the mobility of the particle is larger than predicted by the Stokes equation for viscous resistance. This is due to the changing nature of the interaction between the gas molecules and these small particles. This phenomenon is known as "slip" and it denotes a lessening of the viscous resistance of the medium as the particle size decreases. EPSTEIN[2], KNUDSEN and WEBER[3] and MILLIKAN[4] have reported expressions dealing with the slip correction in terms of the particle mobility Z. The KNUDSEN and WEBER equation is applicable to particles of any size but inappreciably affects particles as large as 10 μ dia. unit density:

$$Z = \frac{1 + (2\lambda/d)\{A + B \exp(Cd/2\lambda)\}}{3\pi\eta d}$$

where λ = mean free path of air, A, B and C, are empirically determined "slip" constants with the following approximate values: 1.25, 0.40 and 1.10, respectively, d = particle dia., η = viscosity of air and 3π = shape factor for spheres.

Another more widely encountered correction factor which is applied to the Stokes equation when dealing with particles approaching the mean free path dimension is the CUNNINGHAM factor $1 + 1.7\ \lambda/d$.[5] The difficulty in using the CUNNINGHAM factor is due to the fact that it does not give a sufficiently accurate correction except over a very limited range, approximately 0.1–0.5 μ dia. Despite this limitation, the CUNNINGHAM correction is widely used. Unquestionably the KNUDSEN and WEBER equation is preferable but has not gained sufficient recognition and use.

In the region of 20 μ dia., particles of unit density will fall with a terminal velocity of about 10^4 μ/sec. At 1 μ dia., the velocity is

* See Ref. 52.

only 35 μ/sec. Despite the fact that very small particles require large slip corrections, they still exhibit extremely low settling velocities, e.g. approximately 0.04 μ/sec for a 0.01 μ dia. particle.

While the size of the particle is the most significant factor in sedimentation, the density of the particle is frequently very important. This is particularly the case in studies involving the heavy metals, their oxides and salts. It is not unusual to encounter conditions in which dusts with a density of 9–11 are involved.[6] A 0.5 μ particle with a density of 10 has the sedimentation characteristic of a unit density particle of about 2 μ dia. As will be seen in the later discussion, this increase in settling velocity may result in an important change in dust deposition.

Sedimentation as discussed here has been limited to the ideal circumstances of tranquil air. There are techniques of approaching the circumstance of stirred settling, turbulent mixing, etc. These are available in many source books.[1,7–9]

B. *Inertial impaction*

Another variation of the basic Stokes equation is utilized in the determination of forces imparted to a particle in an air centrifuge. In this situation, the force of gravity is replaced by the angular velocity imparted to the particle and the radius of curvature involved.[8] This requires, however, that the Reynolds number be very low (less than 0.05) so that the Stokes relationship is still applicable. The Reynolds number for the particle is equal to the ratio of the particle inertia (density $\times$ diameter $\times$ velocity) to the viscosity of air; therefore, the resistance offered by the medium to the particle can change with different particle velocities. Just as the relative motion of the particle to the air is significant, the air velocity and the size and shape of any obstacle are important in governing the flow pattern about the obstacle. The air flow pattern in turn will have a bearing on the behavior of the particle with respect to the obstacle.

Generalizing, the impingement of particles on to a surface from an air stream is possible whenever the inertia of the particle is sufficiently great to cause it to overcome the resistive forces of the medium and intercept the surface or obstacle. Neglecting gravity, the path of the particle in an air stream of changing direction will be determined by the air velocity, the size and shape of the obstacle, and the mass of the particle and its air resistance. Details of the analysis of particle impingement from air streams are often complex and can be found in many references[8,9] but particularly useful are the papers of DAVIES and co-workers[10–13]. These papers, however, do not deal with a model which can be readily transposed to the anatomical configuration of the respiratory tract.

One crucial point in considering inertial impingement in the respiratory tract is whether or not there is a combination of high air flows, particle size and density to produce important inertial effects. Linear air velocities of about 150 cm/sec regularly occur in the trachea. About 200 cm/sec air flows are estimated to occur in the major bronchii and their first branches. These velocities suggested to LANDAHL[14] that Reynolds numbers for air in these regions would be around 100 and possibly as high as 1000. These Reynolds numbers indicate that intermediate or near turbulent flow conditions normally exist in the respiratory tract and turbulence probably exists in these regions during exercise and rapid breathing. A dominant feature of high velocity air flow is that it more nearly approaches an ideal fluid without viscosity.[8] Even so, Stokes' Law for the viscous resistance of the air with respect to the particle is unaffected. Calculations of air flow characteristics at high Reynolds number have been made by LANDAHL and HERMANN[15] and by RANZ and WONG[16] and these are presumably applicable to respiratory tract conditions.

LANDAHL estimates that in a bifurcating tube of order a, the impaction efficiency (fraction of particles removed) will approach 50 per cent when the following expression equals unity

$$\sigma d^2 V_{a-1} \sin \theta / 18 \eta R a,$$

θ is the angle between axes of tubes of order a and a–1, V the average velocity of air stream, d the diameter of particle (sphere), σ the

density of particle (effective; $\sigma-\rho$), η the viscosity of air and Ra the average distance a particle must pass perpendicular to the air stream in a tube of radius R.

However, the problem of determining the average velocity of air flow in various parts of the respiratory tract is difficult, as there is a complex distribution of velocities. Not knowing whether to treat the distribution as rectangular or parabolic, LANDAHL derived expressions for both, and used the average velocity value.[14] From such an analysis, LANDAHL has estimated the role of inertial impaction on dust deposition at various levels of a stylized respiratory tract during a stylized respiratory cycle. These estimations indicate the importance of inertial impaction on the nasal-pharyngeal and tracheal-bronchial areas particularly for particles above about 5 μ dia. (unit density). Dust particles of this dimension have quite appreciable settling velocities (8×10^3 μ/sec) owing to gravity. Since their average residence in the bronchiolar tree might be about 2 sec (respiratory phase time) and the average distance from the center of the air stream to an epithelial surface is on the average no more than 5000 μ even at major bronchii, one would surmise that one important effect of impaction would be to shift the deposition site toward the nasal-pharyngeal region rather than to increase the almost certain deposition of these particles.

As in the case of sedimentation, the inertial force imparted to a dust particle will increase if the particulate density is increased and this again will tend to shift the deposition of dust to an earlier site. It should be remembered that all inertial equations for small particles at low Reynolds number should include a "slip" correction factor.

C. *Brownian motion*

Particles below 0.1 μ dia. (1000 Å) show an increasing susceptibility to random motion imparted by air molecules, known as Brownian motion, although gravitation is still demonstrable, for example, by analysis of the distribution of particle sizes as a function of the height above the earth.

Particles undergoing Brownian motion are characterized by a series of relatively high velocity excursions of relatively short duration in random directions. The displacement of particles in a direction, x, is denoted by the value of the mean square displacement of the particles $\Delta\bar{x}^2$, during time, t, in accord with the EINSTEIN equation[17]

$$\Delta\bar{x}^2 = 2Dt \cdot (S_c)$$

where D is the diffusion coefficient and S_c is the slip correction factor. The diffusion coefficient for spherical particles D is equal to $k \cdot (1/3\pi\eta d)$, where k is the kinematic gas constant, η the viscosity of air and d the particle diameter; therefore, D is equal to k times the particle mobility Z (see above). Note that the density of the particle is not involved.

If a particle of high mobility (small d) is placed in a system where particles are only a few microns from a surface in any direction (small x) and the time is prolonged (large t), the likelihood of particle deposition will be high, $x \alpha \sqrt{t/d}$. Another useful form of the diffusion equation states that the net transfer of particles, P, on to or across a perpendicular plane will be $P = (1/2)(\Delta\bar{x}^2)\ dn/dx$ where $n =$ the number concentration of particles.

An interesting experimental device has been described[18] which makes use of the differences in diffusion capacity of particles of different size. Since it involves a series of closely packed parallel plates, it has been called a "diffusion battery". DEMARCUS and THOMAS[19] found that the fraction of particles of a given size passing through a given battery was a function of the transit time and the diffusion coefficient of the particle. They investigated particles from 200 Å to 6000 Å dia.

If a equals the separation of the plates and b and z their height and length, respectively, and Q the flow through the system, then F, the fraction of particles of a size that emerged from the system, was found to be proportional to $K_1 \exp(-\alpha) + K_2 \exp(-11.4\alpha)$ where $\alpha = K_3 Dbz/Qa$, K_1, K_2 and K_3 were found to have the values 0.91, 0.05 and 3.77, respectively, and D is the diffusion coefficient of the particle involved.

As Q changes in a given structure of a, b and z dimensions, then the transit time changes accordingly. If one can measure a value of F

then it is possible to solve for D. Actually D is best determined graphically by extrapolating to zero flow because the flow is not precisely laminar in such an experimental system.

It is also interesting to note that the DEMARCUS and THOMAS equation is substantially that of TOWNSEND'S[20] who proposed an equation for diffusion in a circular tube during laminar flow.

In the lower respiratory tract one can envision a tremendous assembly of diffusion batteries of essentially circular cross-section. Consider the terminal bronchioles, for example. It is believed that there are between 5 and 6×10^4 of these, each about 0.3 cm long by 0.06 cm dia. Distally to these structures is an even greater number of finer tubes approaching alveolar dimensions (0.02–0.03 cm). From this picture of lung architecture one can imagine the high probability of deposition of any particle reaching this region.

FINDEISEN[21] estimated, using a model lung and respiratory cycle, that the deposition of particles 0.03 μ dia. would be 55 per cent complete in the parenchymal region. Many experimental studies such as those by DAUTREBANDE[22] and VERZAR[23] indicate a much greater deposition, 70–100 per cent for submicronic sizes. An interesting observation was reported by WIGAND[24] who found, with a nuclei counter, that the air he breathed contained about 25,000 nuclei/cm^3 in the inspired air but only 1350 nuclei/cm^3 in the expired air.

D. *Other modifying factors*

1. *Coagulation.* There are several other phenomena of interest which are associated with submicronic particles. Besides interacting with surfaces, small particles interact with other particles, a process known as coagulation or aggregation. According to the studies of WHYTLAW-GRAY and co-workers in the early thirties,[25] the number of (smoke) particles, per unit volume of air, changes with time at a rate proportional to the square of the number concentration n^2; viz. $dn/dt = K'n^2$, where K' is a constant for a given sized aerosol and is larger the smaller the particle size, (K' includes a slip correction factor). Since in a coagulating aerosol the particle size is continuously increasing, the values of K' and n are also continuously decreasing.

Experimentally, it has been shown that the coagulation rate of a polydisperse aerosol can be faster than that of a monodisperse aerosol. This is a function of the ratio of the particulate radii involved and of their absolute values. It has also been reported that the effective radius for coagulation is possibly 10–20 per cent greater than the geometric radius of the particle, owing to intrinsic forces such as Van der Waals' forces.[26] Quantitation of these effects is complex and at best approximate; however, it is interesting to note that they are in the direction of increasing the instability of aerosols.

2. *Electrical charge.* Nearly all aerosols produced in the laboratory by grinding, aspiration, atomization and combustion are found to be electrically charged. Usually the net charge of the aerosol is zero but under certain circumstances may be predominantly of a single sign.[27] In any event, the presence of charges on particles would be expected to induce additional properties in them.

The work of KUNKEL[28] and of DALLA VALLE and co-workers[27] indicates that, in general, only a few electrical charges per particle occur. Condensation aerosols, such as those produced by the La Mer generator, show negligible initial charge but may then accumulate charge by the diffusion of gaseous ions. Therefore, one may conclude that charged particles are a natural phenomenon.

The question then arises as to the possible effects of such charging. One could predict that the attractive and cohesive forces between particles, the so-called effective radius for coagulation, would be increased; the vapor pressure of small particles would be reduced, thus affecting both condensation and evaporation processes[9] and that the migratory movement of particles would be increased in an electric field. Despite the fact that the effect of charges has not yet been fully assessed for each of these phenomena, it is probable that the over-all effect is unimportant except for charge coagulation, and this too is probably minimal owing to the small number of charges involved. Certainly, in animals there is no need to consider the effect of an electric field as none exists

within the airways of the respiratory tract except that due to the charged particles themselves.

3. *Condensation.* The role of submicronic particles as condensation centers was mentioned in the preceding remarks. Hygroscopic particles can initiate condensation at relative humidities well below saturation, e.g. 70 per cent.[29] SCHIFF *et al.*[30] have reported that among a group of inorganic salts, their relative efficiency as condensating nuclei for water could not be correlated to their solubility, crystallization or hygroscopicity. It is well known that there is a critical lower size limit for self-nucleation and it ordinarily requires high supersaturation (4 times or more) for condensation to be initiated without nuclei present.[31] In addition, it is generally stated "insoluble" or nonhygroscopic particles may be important only as condensation nuclei where saturation or supersaturation prevails. Yet the role of such particles and the possible mechanisms involved is clearly not established. For example, they may adsorb a monolayer of the vapor phase and then proceed to behave as though they were a droplet form of the vapor. Whether or not it is important for the particle to be isomorphic with the condensed solid phase is still questioned, and the probable role of microcapillaries in the particle surface[32] is not established. LANDSBERG[33] reported that continental aerosols are composed of both soluble and insoluble nuclei and WAIT[34] has indicated that insoluble nuclei are important in natural condensation phenomena.

While the electric charge, chemical nature and radius of curvature of the particle are among the factors influencing the initiation of condensation, the principal factor governing the rate of growth of small particles is diffusion. This growth process under ideal conditions is isothermal but under normal conditions the growth rate frequently deviates from the predicted, presumably owing to such factors as turbulence and temperature changes.

All direct experimental evidence uniformly implies that submicron hygroscopic particles will grow in size within the water-saturated (or near saturated) air of the respiratory tract.[35,36] The ultimate effect of this growth will depend principally on the size of the particles involved and their residence time within the respiratory tract. It is conceivable that some small particles may actually grow to a more stable size while others will acquire sufficient mass to become more unstable and sediment more rapidly and completely.

4. *Thermal effects.* In the environment, thermal gradients play an important role in the distribution and deposition of dusts. As SAXTON and RANTZ[37] have shown experimentally, the thermal force is proportional to the temperature gradient and the particle diameter. The differential bombardment of the particle by gas molecules produces a motion of the particle towards a cooler area. If the thermal conductivity of the particle is low, an unequally heated particle may result. This, in turn, induces a radiometric effect in which a layer of gas molecules "flows" from the heated side thereby creating an additional force to move the particle towards the cooler region.[38]

In the respiratory tract, if a thermal gradient exists, it is low grade, short-lived and would tend to be repulsive rather than attractive as the particles and the air would ordinarily be cooler than the mucosa of the tract. After the inspired air reaches temperature equilibration, the resulting increase in the absolute temperature ($\sim$10 per cent) would be expected to produce some slight increase in Brownian motion phenomena. In both instances, it is probable that thermal effects on dust deposition are of minor importance in the respiratory tract.

5. *Dust concentration.* References are frequently made to studies[7] which relate the concentration of dust breathed to the amount of dust deposited in the respiratory tract of subjects. For example, in one study as the dust concentration was raised from 3 to 80 mg/m^3, the dust deposition was reduced by 50 per cent. The inference is that as the mass concentration rose, the amount of dust respired decreased. This would follow if (a) the number of very large particles ($>50\ \mu$) was increased concomitantly or (b) if the number concentration of all particles was increased appreciably or (c) if both conditions occurred together. In the first instance, it is presumed that the dust could be sampled quickly enough to sample the larger nonrespirable particles. These particles would

effectively determine the mass of dust sampled while not representing a true dust concentration with regard to respirable sizes. In the second case, the rate of aggregation would be greatly accelerated creating the same general picture; viz. a larger non-respirable particulate population.

One can imagine circumstances wherein the measurement of the mass of dust per unit volume of air might represent any of a variety of particle size distributions. Consequently, a change in mass concentration might be expected either to decrease, increase or not change the deposition, and all of these cases have been encountered.[7] This statement is made with respect to the measurement of the *percentage* of the aerosol mass inspired that is deposited, but frequently applies to the *absolute* amount of dust deposited.

The two parameters which must be analyzed to estimate the effect of altering dust concentration are the particulate concentration and the particle size distribution. However, these must be analyzed in terms of the *time* of the exposure since the system is undergoing active aggregation and sedimentation, causing the dust being breathed from moment to moment to differ.

It is reasonable to generalize that to increase the mass concentration of an airborne dust is to increase the instability of the dust. Since the major changes produced would involve the loss of small particles by aggregation and the loss of large particles by sedimentation, one would predict that the respiratory tract deposition picture would shift to favor dead space deposition, though reduced. Ultimately it would be expected to reflect the deposition of an aerosol, with much less heterogeneity, i.e. principally consisting of particles of the order of 0.1–0.5 μ dia. Then, if the dust ages a few minutes before it is respired, the particulate concentration will probably be less than 10^6 particles/ml air.[25] These aging effects would not be discernible, however, if the dust was being generated continuously.

E. *Discussion*

This brief view of the various physical forces involved in dust deposition suggests that in the respiratory tract the dominant forces are (a) gravitational settling for the large particles (>0.5 μ dia.) and (b) Brownian motion impaction for the smaller particles (<0.1 μ dia.). Small particles may also be deposited by sedimentaton following aggregation with large particles. Since qualitatively the same forces and circumstances prevail in the environment as prevail in the respiratory tract, one can conclude that between 0.1 and 0.5 μ dia. is a region of particle size which is least affected by combined Brownian motion and gravitational forces, and therefore should constitute the most stable aerosol configuration. Deposition data from human and animal studies support the viewpoint that a smaller percentage of particles in this size range are subject to deposition.[39,40] This particular size region is often referred to as the size for minimal deposition. There is a growing misconception that this represents a region of particle size of relatively less importance to deposition than any other. This is traceable, in part, to particle size deposition relationships such as reproduced in Fig. 1.[21] (See also Fig. 3.)

These data are based on the percentage of particles of a given size being deposited (retained); however, there is no indication of the relative or absolute numbers of such

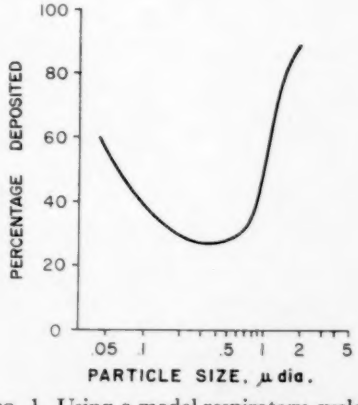

FIG. 1. Using a model respiratory cycle and respiratory tract, and by adding the contribution of sedimentation, inertial impaction and Brownian motion at each particle size at various anatomical levels, Findeisen was able to construct the deposition curve shown. Unit density spheres were the particles assumed.

particles that may be involved. In aged aerosols, since this 0.1–0.5 μ dia. size region is more stable, greater numbers of these particles will probably be present to be breathed. Even in freshly prepared aerosols, the relative numbers of particles in this region may be orders of magnitude greater than those above 5 μ. If one considers a factor of 5 as encompassing the variability of particles of different size in terms of deposition (20–100 per cent), then the relative and absolute greater number of 0.1 μ dia. particles deposited may greatly exceed the number and even the mass contribution of particles $\sim$5 μ dia. which are deposited.

In many toxicological situations the hazard is proportional to the mass of the aerosol deposited. When this is so, the largest particles penetrating the respiratory tract are probably the most significant. This is unquestionably the case with a readily soluble or absorbable material as was so aptly demonstrated by PUNTE[41].

If the significance of the exposure is based on the penetration and retention of relatively insoluble material in the parenchymous areas of the lungs, the fate of the large particles (>5 μ) is probably of little consequence. Moreover, the relatively high deposition and retention of small particles (<1 μ) in the lung parenchyma has been repeatedly demonstrated.[22,23] This suggests that a measurement such as the specific surface area of the dust may prove to be a better index of its retention (solubility) and of its propensity to produce localized toxic reactions than does its mass. Such a circumstance was presumably encountered in one investigation involving oxides of beryllium, wherein their acute toxicity was apparently related to their specific surface areas as determined by gas adsorption.[42] In addition, there is evidence that airborne particles may serve as vectors for the admission of toxic vapors,[43] micro-organisms[44] and radioactive atoms[45] into the respiratory tract. This probability also suggests the importance of surface area or other parameters than mass, thereby adding to the significance of small particles.

To decide which particles are the most significant in assessing the hazard of a given dust exposure consequently requires some knowledge of the toxicology of the material. Such matters as solubility, the response of tissues to the "insoluble and soluble" forms of the chemical moiety, some appreciation of the dose-response characteristics of the material and species involved, etc. are often decisive. The more physical, chemical and biological data available, the more likely that a correct judgement can be made regarding the dust exposure.

These facts do not reduce the attention which submicron particles deserve in toxicological and particularly in radiological situations. This attitude is justifiable for two important reasons, (a) the knowledge that these particles are indigenous to all environments of man and (b) the overwhelming evidence that they have an inordinately high deposition and retention, and more often than not, prove more toxicogenic.

Whatever dust sampling devices are used for the evaluation of dust hazard, they should be evaluated in terms of their ability to measure the small particles as well as the large. This, in turn, implies further requirements for particulate analysis, such as electron microscopy as a routine.

In most inhalation studies, estimates of mass and surface area distribution, etc., are obtained by first determining the size-frequency distribution.[7,9] Specifically, if a particulate population follows a commonly found frequency–size distribution, it can be normalized using the logarithm of the particle size. After being normalized, the medium size and standard deviation of the distribution can be determined. With these two parametric functions, various other relationships of the aerosol, such as surface and mass, are predictable and may be related to the size distribution. A typical size and mass distribution is plotted in Fig. 2.

Without the benefit of such a statistical treatment, the determination of these various relationships is complex and at the present time, not technically feasible. Without these relationships, many deductions based on particle size are untenable. It is obvious that the phenomena of interest, especially sedimentation and Brownian motion, are so size-related as to demand the consideration of particle size as the primary *physical* factor affecting deposition.

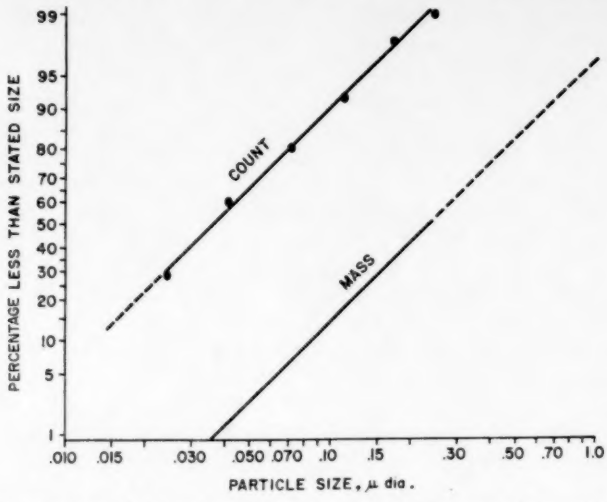

FIG. 2. In this particulate distribution approximately 40 per cent of the mass resides in particles between 0.1 and 0.25 μ dia. Although no particles were seen or measured in excess of 0.2 μ the distribution predicts that about 0.001 per cent of the population will be larger than 0.3 μ and these few particles will also contribute about 40 per cent of the mass of the system. On the other extreme, note that even though 90 per cent of the particles in the aerosol are less than 0.1 μ they contribute only about 15 per cent to the total mass.

III. PHYSIOLOGICAL FACTORS

The air of the respiratory tract cannot be simplified into a static compartment of warm, moist air. The ability of the lung air to change in composition, volume and aerodynamics continuously, brings additional factors into play which also affect dust deposition.

The composition of respiratory tract air changes continuously, chiefly because it is unevenly and periodically ventilated. Among other things, this implies that in different parts of the lung, the particles of dust inhaled will have different residence times and that deposition will not be uniform.

A. *Effects of tidal volume and respiratory frequency*

The volume of the respiratory tract varies about 15 per cent with each tidal volume. Approximately one-third of this air, the so-called dead space air, can be considered to reside in the respiratory tract for the duration of one respiratory cycle. This will also be the case for dust suspended in this air volume, except for that dust deposited during the cycle time. If the volume of the respiratory tract is periodically increased (by deep breathing) the number of dust particles breathed is increased and the depth of the tract which they reach is also increased. If the respiratory frequency is decreased concomitantly, the mean residence time of the particles is increased—even those in the dead space air, because the cycle length is prolonged. This combination of increased volume and time would be expected to increase deposition on theoretical grounds and this expectation has been repeatedly verified by experimentation.[40,46]

On the other hand, when the respiratory frequency is increased, the mean residence time for the particles in the anatomical dead space should decrease. Depending on the tidal volume, there should be an increase in air turbulence and in particulate velocities. Increasing the air and particulate velocities would be expected to produce an earlier deposition by impaction and possibly a greater deposition owing to increased inertial impaction and turbulence. These tendencies in the face of decreased residence times for particles create a picture which is difficult to analyze.

Several attempts to relate deposition to respiratory velocities have been reported. In

one study,[47] with human subjects breathing normally, an increase in mean inspiratory velocities was reasonably well related to an increase in mass deposition among subjects. Utilizing the same submicronic aerosol in dogs[48] the same relationship appeared to hold. However, the effect of expiratory velocities on deposition was not consistent in the two studies. Some observations of DENNIS[49] suggest that there may be variable effects of respiratory frequency (respiratory velocities?) on deposition. In DENNIS' experience at both lower and higher than normal breathing frequencies, deposition increased while it was minimal at the middle frequencies. It can only be speculated whether, at high respiratory frequencies, this increase is an expression of an air velocity effect.

Since the exchange of gases at the alveolar level is normally controlled by diffusion, small particles entering the alveolar air will be relatively less influenced by the usual respiratory excursions. Deep breathing, wherein the alveolar air is more directly exchanged, should increase the penetration of dust into the lung air keeping the particulate concentration high, thereby enhancing diffusion into alveoli. Therefore, it is probable that changes in respiratory frequency and volume will produce changes in both the site and degree of dust deposited throughout the lung, chiefly by increasing the mean residence time of the particles and the number of particles involved. Creating better mixing and greater particulate inertia by rapid breathing may be effective in increasing deposition and particularly, for shifting the sites of deposition, but interrelations with other factors, such as shortened residence time, suggest these latter phenomena may be more effective in altering dead space deposition than alveolar deposition.

The foregoing picture is basically an analogy of the lung ventilation process. Let V_A equal the alveolar ventilation (volume/unit time), F the respiratory frequency, V_T the tidal air volume and V_D the anatomical dead space volume*; then $V_A = F(V_T - V_D)$. This implies that if one breathes with 6 liter min volume and increases the respiratory frequency from 8 to 24 cycles/min while decreasing the tidal volume from 750 to 250 ml, one's dead space ventilation (total ventilation — alveolar ventilation) will be increased four-fold.

When one considers the intimate relationships involved in respiration, it becomes extremely difficult to isolate primary determinants. As already cited, in investigating the effect of an increasing tidal volume at a constant respiratory frequency, the question arises as to which of the volumetric or dynamic changes involved are responsible for the observed effects on deposition.

Despite the difficulties involved, there have been many experimental studies[39,40,46,47] reported which demonstrate that the amount of dust deposition occurring is greatly affected by the character of respiration. The amount of change in deposition that can be induced is at least comparable with that produced by different particle sizes. The following graph (Fig. 3) summarizes many of the important inhalation studies in which some recognition was given to the physiology of respiration.

From these data we can see, for example, an effect of changing from slow to fast respiration

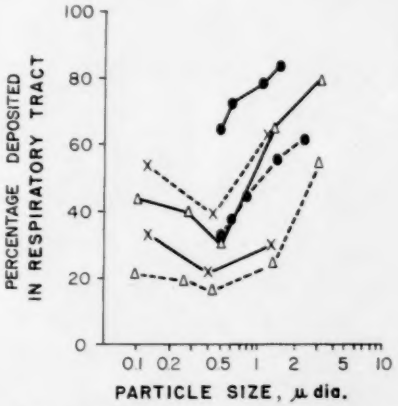

FIG. 3. WILSON and LAMER[40], 5.5 cycles/min (closed circles and line), and 20 cycles/min (closed circles and dashes); LANDAHL[14] (and co-workers), 5 cycles/min (triangles and line), and 15 cycles/min (triangles and dashes); ALTSHULER *et al.*[46] (Subject P) 9 cycles/min (crosses and dashes), and 21 cycles/min (crosses and line).

* Unlike the functional or physiological dead space, the anatomical dead space is a nearly constant volume ranging from 150 to 170 ml.

on the deposition of 0.5 μ dia. particles (WILSON and LAMER) was to decrease the percentage deposition from approximately 70 per cent to approximately 35 per cent. In LANDAHL's study, changing the tidal volume and respiratory frequency to maintain a constant minute volume resulted in qualitatively the same effect on the deposition of 0.5 μ particles but not to the same extent. However, in the case of 1 μ particles, the percentage deposited was more than doubled when the respiratory frequency decreased from 15 to 5 cycles/min and the tidal volume increased from 450 to 1350 ml.

There are obviously many other volumetric and aerodynamic parameters that can be measured during respiration. Among these there may be significant relationships to deposition, but at present, this is pure speculation. Also related to the deposition phenomenon are certain effects induced by dusts on the respiratory physiology. DUBOIS and DAUTREBANDE[50] have shown that the respiration of insensible amounts of dust causes a decrease in lung compliance. This effect is one which involves the constriction of the fine air tubes and possibly parenchymous structures. While this response must be considered normal and not discomforting to the subject, it may have a considerable bearing on the ensuing dust deposition pattern. This constrictor effect of dusts is also being studied as a possible factor in the genesis of pulmonary dysfunction and disease.

B. *Discussion*

To date, most physiological relationships in dust deposition have been obtained empirically. Although some attempts have been made to predict deposition as a function of physiological measurements, these formulations so far are unpretentious and admittedly inaccurate. It has been necessary, however, for certain "Committees" to create formulations upon which they hope *conservative* estimations of hazard can be based. An outstanding example of this is the so-called Harriman Conference model[51] which states that standard man will breath 2×10^4 l. of air each day and will deposit 75 per cent of the dust he breathes.

Table 1

Factors	Particles involved	Related factors	Primary effects	Special considerations
Sedimentation	>0.1 <50 μ	Particle shape and density Particulate concentration (aggregation), "slip", hygroscopicity	To cause dust deposition in nasal pharynx and and tracheal-bronchial tree	Highly significant in toxicological studies, probably the most important depositon factors. Especially significant with "soluble or absorbable" dusts
Inertia	>0.5 <50 μ	Same as above plus relative velocities of particle and air	Same as above with tendency to promote earlier deposition	
Brownian motion	$>0.002(?)<0.5$ μ	Electric charge, hygroscopicity, particle concentration (aggregation), thermal gradients	Dust deposition where large and intimate surfaces involved, viz. lung bronchioles and and parenchyma	Most important when dust is insoluble and submicronic Also significant when these dusts serve as vectors
Respiratory frequency	All sizes	Air velocities, residence times, turbulence, dead space ventilation	Increasing tends to *decrease* deposition (possibly very high rates e.g. >20 may increase deposition	Exercise, physical labor, and heated environs will have variable effects on subjects but, in general, will tend to increase both these factors together
Tidal volume	All sizes	Number of particles, alveolar ventilation, residence time	Increasing tends to *increase* deposition (even at constant minute volume)	

Of this only one-third will be deposited in the lower respiratory tract (i.e. 25 per cent). In addition, the lower respiratory tract retention is presumed to follow a pattern whereby one-half (12.5 per cent) of the dust will be redistributed in 24 hr and the remainder retained with a half-life of 120 days (insoluble dust). The estimate of 2×10^4 l. of air breathed per day and the over-all deposition figure of 75 per cent are conservative ones and would not be expected to underestimate an exposure. However, the lower respiratory tract deposition value of 25 per cent and the clearance rates suggested, could conceivably misjudge an inhalation exposure to the lower respiratory tract to an important degree. Recalling, for example, VERZAR's[24] work in which dogs breathed a submicron aerosol through their nose and through a tracheal cannula, it was found that the upper respiratory tract contributed only a small amount, viz. it reduced the over-all dust deposition from 77 to 64 per cent.

The property of selecting certain clearance rates is beyond the scope of this discussion but reference is made to Part II of this paper (CASARETT[52]) to the NAS–NRC Inhalation Hazards Subcommittee[53] and Committee on Pathologic Effects[54] reports.

IV. SUMMARY

A brief summary of the various physical and physiological effects discussed is given in Table 1.

At present it is not possible to predict a great deal about a dust exposure. Retrospective analysis is often more fruitful. Even then, we obtain only an approximate picture, for deposition is inherently concerned with more than the amount of dust invading the respiratory tract—but also the site of the deposition and the physical–chemical nature of the deposited dust; therefore, the deposition process is inseparably related to, if not a principal phenomenon governing, retention in the respiratory tract.

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SOME PHYSICAL AND PHYSIOLOGICAL FACTORS CONTROLLING THE FATE OF INHALED SUBSTANCES—II*†

RETENTION

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Abstract—Some of the mechanisms by which particles are cleared from the lung are discussed. Emphasis has been placed on relatively insoluble particles deposited in alveoli. The fundamental mechanisms of clearance are discussed, with some emphasis on those aspects which are pertinent to the calculations of the radiation dosage to the lung as usually performed.

A. INTRODUCTION

PARTICULATE material deposited in the respiratory tract behaves in a variety of ways. The following is a brief review of certain aspects of the behavior of particles in the respiratory tract. Emphasis is placed on relatively "insoluble" particles, deposition in the parenchymal regions of the lung and on phagocytosis and its part in the mechanisms by which the particles are removed from the lung. In addition, some emphasis is placed on the significance of these mechanisms in estimating dosage delivered to the lung.

The retention of material by the lung, i.e. the persistence of deposited material in the organ, varies greatly. Whether a material is "cleared" slowly and has a long residence time in the lung or is rapidly cleared from the lung, the rate of clearance observed is a resultant of the interplay between the physicochemical properties of the material under study and the physiological responses elicited. These two sets of factors are interdependent. However, for purposes of the present discussion of the physiological mechanisms of particle clearance, the physical and chemical factors are viewed only as they influence these mechanisms.

The nature of the material deposited can influence clearance directly or indirectly. An example of a physical characteristic directly affecting clearance is solubility. A soluble material would be expected to be absorbed rapidly from most surfaces within the respiratory tract thereby obviating the necessity of other physiological processes. Indirectly, the nature of the material also influences the modes of clearance in a variety of ways. Of primary interest in the present discussion is the influence of the particle characteristics, particularly particle size, on the site of deposition as discussed by MORROW[1]. The site of the deposition determines the morphological entities which are available for interaction with the particle. This interaction, in turn, establishes the mode of removal of the material from the lung.

Finally the physiological response (and therefore the rate and mode of clearance) depends upon the functional state of the tissue components which are brought into play. Among the variables are such things as differences in affinities between particles and phagocytic cells, differential response to particle size, deleterious effects of the material inhaled, or pre-existing pathological conditions. In short, the physiological response depends upon

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the fundamental relationships between cell and particle, and upon the degree to which the functional integrity of the tissue remains intact.

Following the deposition of relatively insoluble particles in the lung, the clearance of the material is classically attributed to several possible routes. Some of these are fairly well established; others are more controversial. If particles are deposited at or above the level of the upper third of the terminal bronchiole, they follow a well-established route of clearance. At this level of the respiratory tree, particles encounter a ciliated mucous epithelium. The ciliary action moves particle-laden mucous up the respiratory tree to the pharyngeal region from which area it is swallowed or expectorated.

If, on the other hand, particles are deposited distal to the respiratory bronchiole, cilia are not present and one must invoke other mechanisms to explain the removal of particles from these sites of deposition. Before passing to a discussion of mechanisms by which particles are moved, one may list some possible fates of particles deposited in the lung.

B. FATE OF INHALED PARTICLES

Particles deposited on the ciliated mucous epithelium are usually cleared to the gastrointestinal tract. From the latter site, some absorption might occur into the systemic circulation. The possibility of penetration of the bronchiolar epithelial lining by particles is discussed below. For particles deposited in non-ciliated, parenchymal regions of the lung, several possible fates may be listed: (1) Particles might persist for relatively long periods of time (weeks to years). (2) Irrespective of the residence time in alveoli, particles might find their way out of alveolar regions to areas of the respiratory tree which have a ciliated epithelium. From such a location, they would be moved up the respiratory tree and out of the lung. (3) Particles might move or be moved to other areas of the lung, e.g. into lymphoid tissue or interstitial regions. Following such movement, persistence in these new locations might vary considerably for different materials. (4) Finally particles might enter the systemic circulation either by way of the lymph or blood vascular system. This might occur from parenchymal or other regions of the lung or from other tissues to which the particles have been previously transported.

Irrespective of what the precise disposition of the particle is, there are several fundamental questions as to how the movement occurs (or why movement does not occur). Bearing strongly on this problem is the question of whether the particles are moved without cellular intervention or whether movement occurs by virtue of phagocytic cells.

The modes of particle movement are discussed in some detail below. The selection of literature is intended to be illustrative rather than comprehensive. Examples of certain facets of the mechanisms of particle movement have been selected from some recent studies dealing with the administration, via the lung, of polonium-210. Complete reports of these studies can be found elsewhere or are in preparation.

C. MECHANISMS OF LUNG CLEARANCE

1. *Ciliated epithelium*

Although cilia have been under investigation from the time of the first compound microscope, much remains unknown about their mode of action and regulation. Many of the theories of action have been obtained from non-mammalian tissue and unicellular organisms as reviewed by GRAY,[2] but it would appear that a reasonable correspondence is found with the ciliary activity of the mammalian respiratory tract.[3,4]

Briefly, the cilia are delicate, tapered filaments projecting from the border of the epithelial cell, about twenty to thirty per cell. The cilia change abruptly from an erect position to a hook or sickle shape and slowly return to an erect position. This movement may be very rapid (12–20 times/sec) and occurs in a co-ordinated way with the complement of cilia from one cell. There is also an apparent co-ordination with adjacent cells to produce wave-like movements (metachronal waves) toward the glottis. In this manner the thin layer of mucous covering the epithelium and particles adhering to this layer are moved up the respiratory tree.

Apart from the ciliary action other factors have been suggested as aids in movement of

particles up the respiratory tree. The widening and narrowing of air passages during respiration may have some effect on the movement of the mucous layer. Distinct from this action, however, is the possibility of a peristaltic wave phenomenon as described by JARRE[5]. Although at least one attempt at confirmation of JARRE's findings has failed,[6] the phenomenon bears further investigation.

Finally the cough mechanism might aid in the removal of particulate matter. Receptors which initiate the cough are present at the tracheal bifurcation but probably not much deeper than this level. Furthermore, the layer of mucous would be expected to prevent stimulation of these receptors by any but large particles, perhaps about 50 μ dia.[7] Thus it is doubtful that coughing contributes significantly to the upper respiratory-tract transport of particles small enough to be deposited in the deeper portions of the lung.

The rate of movement of particulate material deposited on the ciliated epithelium is rapid under normal circumstances. Various estimates of the rate of movement have been obtained; all indicate an increasing rate from the small bronchioles to the bronchi to the trachea. For example it has been found that rates of about 0.15–0.30 cm/min can be expected in bronchioles,[7] 0.25–1.0 cm/min in bronchi[8] and about 3–4 cm/min in the trachea.[7,9] These rates are dependent upon the functional activity of the ciliated epithelium which is, in turn, subject to other influences.[10]

The possibility has been stated that particles might penetrate the bronchiolar epithelial cells or the epithelial layer. Although most investigators would contend that the epithelial cells are non-phagocytic, some evidence has been presented to show that under certain circumstances epithelial cells (in lung and other tissues) take up particulate material. For example, ROPES[11] found the cuboidol, ciliated cells of the terminal bronchioles of the rabbit lung took up inhaled particles. Other investigators have obtained similar results.[12,13] The possibility that particles penetrate the epithelial lining of bronchioles has not been widely espoused. There are, nevertheless, a number of observations which suggest that such movement does occur. A recent example of such an observation is shown in Fig. 1. This is a section of trachea of a rat after inhalation of polonium-210. At all levels of the respiratory tree radiocolloids were found which had apparently penetrated the epithelial layer. It has been suggested[14] that these colloids had been phagocytized and that penetration of the epithelium was accomplished by migration of the phagocytic cell. Although these possibilities are of considerable fundamental interest, there is no body of data which would point to any quantitative significance of bronchiolar penetration with respect to the changing lung burden or to the distribution of the radiation dose.

Of greater significance in dosage calculations is the pattern of the ciliary mucous "tide". In early investigations the path of particles has been described as spiral in nature.[9] In recent years several aspects of ciliary movement have been scrutinized intensively by A. C. HILDING. Some of HILDING's findings have a direct bearing on considerations needed to assess the radiation dose delivered to the tracheo-bronchial tree.

It is well established that the volume of the components increases greatly from the trachea to the alveoli. The bronchiolar tree, however, does not increase greatly in volume or cross-sectional area from the trachea to the small bronchioles[15] as is commonly assumed. Rather one finds about 50–100 per cent increase in volume from the trachea to bronchioles measuring about 1–2 mm in diameter.[15] A much larger increase occurs, of course, in the total surface area of the ciliated mucosa in these regions of the lung. A further increase in cross-sectional area by about a factor of 10 occurs between small bronchioles and alveolar ducts[16] with a concomitantly greater increase in surface area.

It is readily apparent that in transport of particles along the ciliated epithelium, a given quantity of material is encountering a more limited surface as it moves toward the glottis. Thus it would be expected that, simply from the quantity of material moved, the dose delivered to the mucosal surface would increase as one moved up the respiratory tree. The resultant of this factor which would increase dose, and the more rapid movement in the larger airways,

which would tend to decrease dose, has not been assessed.

If the entire surface area of ciliated epithelium is used for calculation of dose to a region, the estimated dose would be too low. This is because the material must be moved around branching bronchiolar openings, thus reducing the surface area over which the mucous flow occurs. HILDING[17] has shown (in the human lung) that at some points the mucous flows over a surface as little as 10–20 per cent of the surface which would be calculated ("ciliary narrows"). For this reason accumulations of particles are often seen at bronchiolar branch points in histological sections (Fig. 2).[14] A similar finding is observed at the deepest part of the ciliated epithelium where accumulations of particles have been observed.[14] (See Fig. 3.)

It has been shown[17] that there are small "whirlpool" areas where material may reside on the epithelium for time periods longer than would be predicted on the basis of the normal rate of movement. These areas would be likely to receive particularly high doses in comparison with surrounding epithelium. The "normal" ciliated epithelium in the human lung often contains small islands of squamous cells. At these regions material being moved along the epithelium tends to accumulate in small pockets[18] thus supplying another possibility for a high local dose to the epithelium.

2. *Phagocytosis*

Perhaps the most fundamental aspects of the mechanisms of lung clearance have to do with the degree and rapidity of phagocytosis of particles in the lung and the origin and disposition of the phagocytic cells. These aspects were reviewed by ROBERTSON[19] in 1941. Since that time more detailed studies of the fine anatomy of the lung have been carried out and, with increased interest in and use of radioactive particles, much additional light has been shed on the interaction of particles and phagocytes in the lung. The underlying physicochemical initiation of this interaction is beyond the scope of this paper. A useful review of this subject is that of MUDD *et al.*[20]

The origin of macrophages throughout the body has been a subject of general controversy. The origin of alveolar macrophages has been especially controversial. Much of the disagreement has been due to lack of knowledge about the intimate anatomy of the lung parenchyma. Early investigators described or supposed a complete epithelial lining of the alveoli which was continuous with the bronchiolar epithelium. Other investigators, including ROBERTSON[19] opposed this view. Irrespective of the view held about the alveolar lining, some authors[21–23] suggested that the general source of the alveolar macrophage was the interstitial tissue and the mononuclear cells of the septa. On the other hand, some adherents to the "continuous-epithelium" view[24–26] suggested that alveolar macrophages arose from septal cells which in turn were derived from the alveolar epithelium.

The latter view has gained wide acceptance in recent years. This is due in great part to the work of LOW,[27–29] KARRER[30–32] and others who have established, among other findings, that there is indeed a continuous alveolar epithelium. In additon, more strength has been lent to this suggestion by recent investigations with radioactive particles and a re-evaluation of the older work in the light of present knowledge.

Many investigations purporting to demonstrate the origin of macrophages in interstitial regions utilized intratracheal injection or the use of materials which were irritative and/or inflammatory and/or infectious. Under such circumstances, it is now recognized that migration from interstitial regions of phagocytic cells, some of which are derived from blood,[33–35] might occur to a degree which could easily mask any contribution of the alveolar epithelium to the pool of phagocytic cells in the exudate. Thus, conclusions based on such studies are not directly applicable to the situation under consideration here. A further objection has been the report that "fixed" cells of the lung, particularly epithelial cells, are not phagocytic.[36–38] However, other authors have reported phagocytosis by alveolar septal cells.[21,24,39,40] Although some very convincing evidence exists against phagocytosis of ThO_2 by alveolar epithelium,[29] it has been suggested that interaction of particle and cell might vary with different materials.[19] The question

remains open to further investigation along the approach described by Low[29].

Following engulfment of a particle, the cell moves up the respiratory tree or into interstitial regions from where it may enter the lymph or blood vascular systems. Several fundamental questions revolve about this phase of particle movement. Direct penetration of the alveolar epithelium into interstitial regions or into the vascular system has been suggested,[41,42] without invoking cellular engulfment. "Emigration" of cells out of interstitial regions, engulfment of particles followed by "immigration" into interstitial regions has been postulated.[43,44] Also unexplained are the reasons why some cells or particles leave alveoli to enter the upper reaches of the respiratory tract while others apparently penetrate the alveoli and are deposited elsewhere in the lung or are carried away to other tissues.

Some recent investigations with Po^{210} have been interpreted in a manner which has some bearing on these questions. Firstly, with regard to direct penetration of the vascular system, some observations tend to suggest that it does, indeed, occur. Fig. 4 is a picture of an autoradiographic manifestation of a Po^{210} radiocolloid found in the circulation with no apparent cellular association. However, this was found after intratracheal injection of the suspension of the colloid.[45] Following inhalation of Po^{210} no such observations were made.[14] It has been concluded, tentatively[26] that, in these studies, the direct penetration observed was an artifact of the intratracheal injection technique and its sequelae.

Comparing the time sequence of events following inhalation of Po^{210} has led to the conclusion that, with this system at least, entry of particles into interstitial regions was not directly through the alveolar membrane, but rather occurred by cellular mediation. Fig. 5 shows a colloid apparently in a lung macrophage. Phagocytosis of the colloid occurred rapidly and 2 hr after inhalation virtually all particles in parenchymal regions appeared to be in phagocytic cells. At this time, very little polonium was found in interstitial regions. On the other hand, 24 hr after inhalation, most of the colloid still present in parenchymal areas appeared to be interstitial (see Fig. 6). Furthermore, whenever unequivocal viewing of the autoradiograms was possible, the colloids appeared to be associated with cells indistinguishable from alveolar macrophages.

The question of why some cells containing engulfed particles move up the respiratory tree while others apparently move to interstitial regions, is unanswered. Recently the suggestion has been made that a plausible answer might lie in the development and renewal of the alveolar epithelium.[46] A number of authors[24,25,39] have postulated the development of macrophages from alveolar epithelial cells and have described the morphological changes during this development. More recently, BERTALANFFY[47,48] has described these changes in more detail and has measured a turnover rate for these cells. If one examines the photomicrographs in these papers and the electron-photomicrographs of Low[27,28] and KARRER,[30,31] it is observed that along with the described changes there is a progressive delineation of the epithelial cells from the underlying tissue. It has been postulated[46] that early forms of the developing macrophage upon stimulation to engulf particles, round up and are moved into interstitial regions by the pressure of surrounding cells. On the other hand, more mature forms in this developmental scheme are more nearly completely separated from the underlying tissue, become detached after phagocytizing a particle and are free to move or be moved up the respiratory tree.

3. *Lymphatic absorption*

The significance of lymphatic drainage from the lungs and serous cavities has been well reviewed by COURTICE and SIMMONDS[49]. The lymphatic uptake of particulate material initially deposited in lung has been widely described and will not be reviewed in detail here.

It is generally agreed[16,50] that the alveoli are devoid of lymphatic channels and that entry of material into the lymphatic system occurs at the level of the alveolar duct or respiratory bronchiole. A particle deposited in an alveolus might take, theoretically, several routes to enter the lymphatic system.

Direct penetration through the alveolar wall

into interstitial fluid is one possibility. The particle would presumably follow movement of interstitial fluid to the origin of the lymphatic system at about the level of the respiratory bronchiole, from which site it would then enter the lymphatic vessel. Contrary to this postulate, it has been suggested[51] that the alveolar membrane constitutes, in fact, a barrier to absorption of proteins and would be, presumably, a more formidable barrier to particulate material. It has been argued[49] that the latter postulate is unlikely because, in pulmonary edema, proteins freely pass from tissue fluids into the alveoli. It would seem that the potentiality of the alveolar membrane as a barrier to direct penetration of particles in normal circumstances should not be dismissed on the basis of observations under pathological circumstances involving altered permeabilities.

In theory, the particle could be moved out of an alveolus to the alveolar duct and respiratory bronchiole and enter the lymphatic system at that point. This has been suggested by MACKLIN[52]. To adhere to the penetrability of the epithelium in this region, and to deny the permeability of the alveolar membrane, one must postulate some "defect" or special structural configuration to explain entry into the lymphatics. No unquestionable evidence for such a structure has been found. On the contrary, LOW[27] describes the continuity of the alveolar epithelium with that of the bronchiolar epithelium.

Whatever the site of entry into the lymphatic system, it is becoming abundantly clear that most material is phagocytized first and the subsequent disposition of the particles depends upon the fate of the phagocytic cells.[49] Although there have been reports of direct penetration of particles without mediation of phagocytic elements,[53,54] it has been pointed out[49,46] that such findings might well be the results of alteration of normal processes following injection of fluid suspensions.

The rate of lymphatic uptake is dependent upon the nature of the material. For example, HAHN *et al.*[55,56] have demonstrated the more rapid uptake of radioactive silver colloid as compared with gold colloid and the increased rate of uptake when the gold was coated with silver. Similarly, it was observed[57] that polonium-210 on silver particles was more rapidly cleared from the lung than polonium radiocolloid alone. At least in part, this was due to more rapid phagocytosis and more rapid uptake by the lymphatic system.[45]

In studies utilizing intratracheal injection of polonium-210 radiocolloid in rabbits,[48] polonium was observed in small masses of lymphoid tissue at the level of terminal bronchioles at 24 hr after administration (Fig. 7). At later intervals after administration, radioactivity was found in lymphoid tissue surrounding larger bronchioles. Thus, although particles leave the parenchymal regions, they remain in the lung for some time before reaching the hilar lymph nodes. Fig. 8 is a picture of particles in the tissue surrounding the trachea. On close inspection these colloids were found to be in lymphatic vessels.[45] The factors affecting the rate of pulmonary flow and thus the rate of movement of particles from the lung by this route have been dealt with elsewhere.[49]

There is little question that the lymph nodes can accumulate large quantities of material. Recent work with inhalation of UO_2 particles[58,59] has shown concentrations as high as 300 mg UO_2 per g of lymph node. It is evident that such high concentrations require some attention to the possibility of damage to these lymphatic organs. PuO_2 is another insoluble material which is of some interest. Earlier work by HAMILTON *et al.*[60,61] indicated that most PuO_2 deposited in the lung was eventually cleared via ciliary action and that no significant role of the lymphatic system was detectable. Although a major portion of the lung load is cleared up the respiratory tract, it now appears that the lymph node accumulation of PuO_2 particles is considerable.[62–64]

The implications of this mode of lung clearance to estimates of the radiation dose are numerous. Firstly, the apparent accumulation of particles at respiratory bronchioles before entry into the lymphatics would be expected to result in a dose considerably in excess of that which would be calculated on the basis of the lung model adopted by the Harriman Conference[65] even if the kinetics of the model were precisely applicable. Secondly,

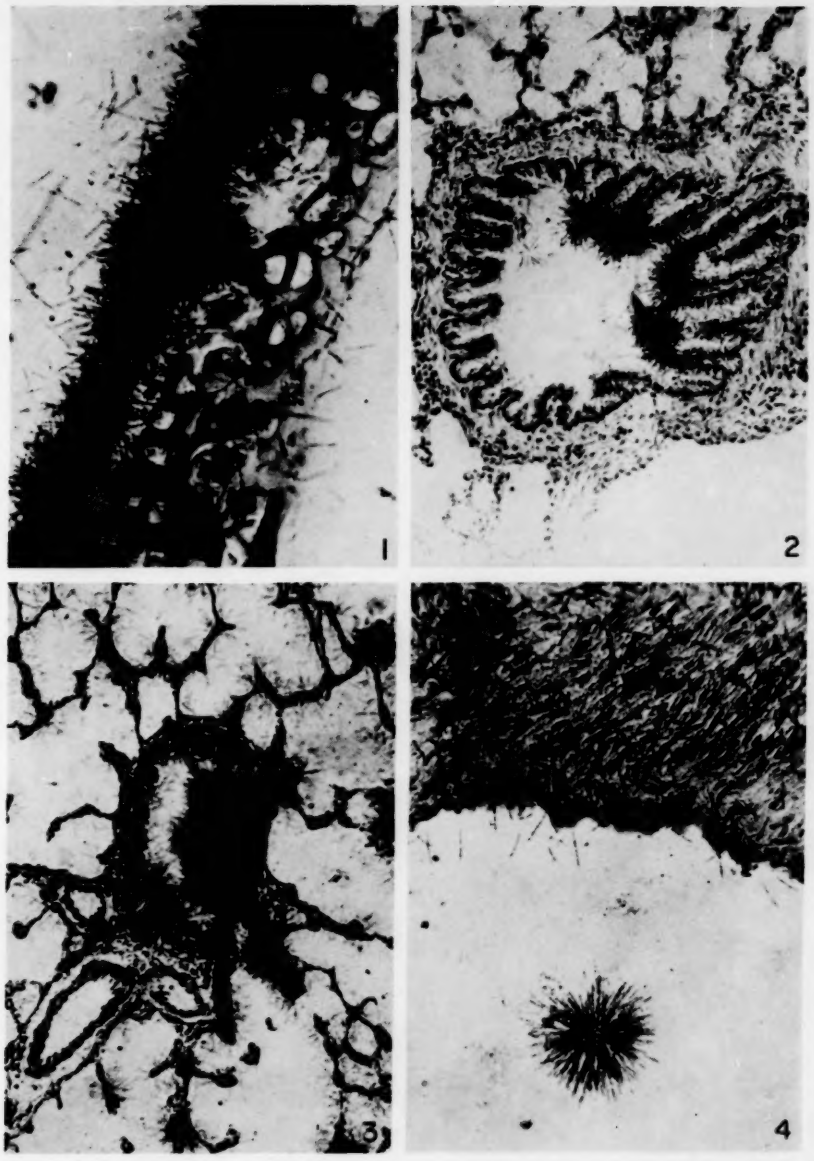

FIG. 1. Autoradiogram of tracheal surface of rat after Po^{210} inhalation. Note apparent penetration of the mucosa by one colloidal particle. FIG. 2. Accumulation of radiocolloids at bronchiolar branch points. FIG. 3. Accumulation of polonium radiocolloids in small bronchiole in deep, parenchymal region of lung. FIG. 4. Polonium radiocolloid in blood vessel apparently unassociated with any cellular element (intratracheal injection).

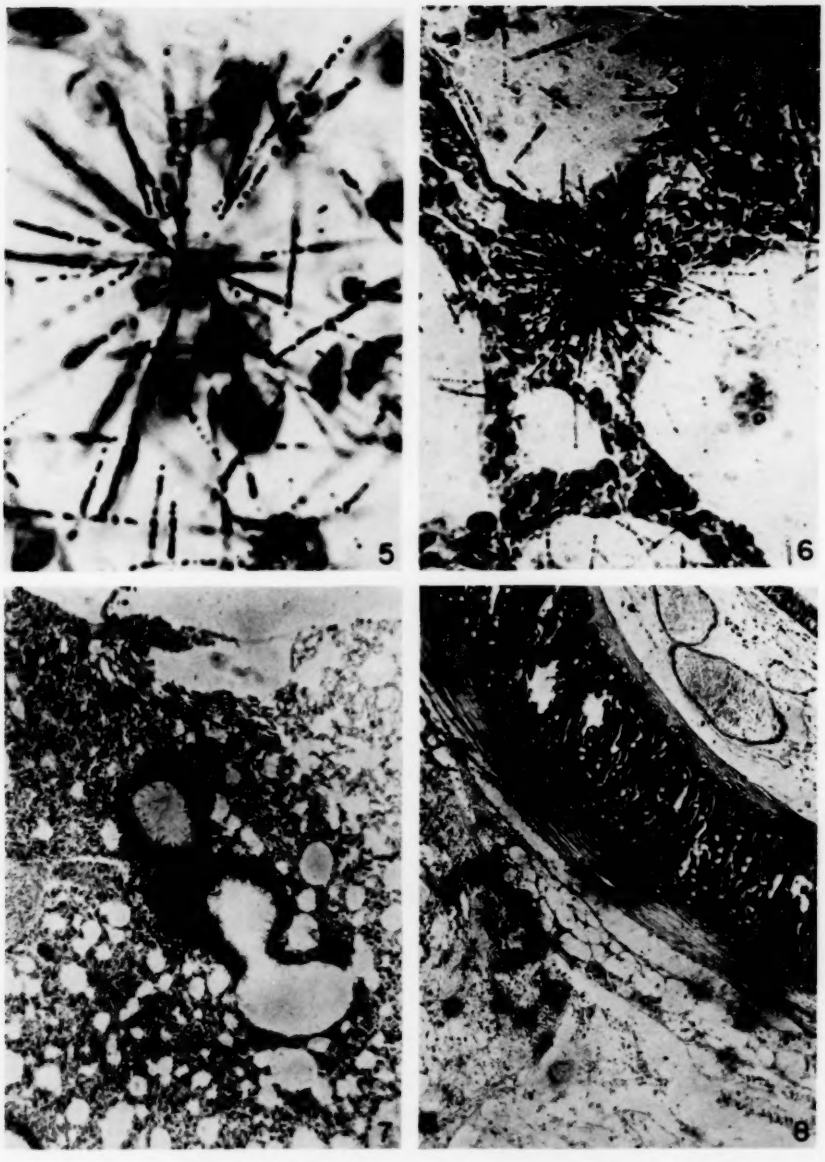

FIG. 5. Polonium radiocolloid in lung macrophage. FIG. 6. Phagocytized polonium radiocolloid in interstitial region. FIG. 7. Polonium uptake by peribronchiolar lymphoid tissue after intratracheal injection FIG. 8. Polonium radiocolloids in tissue surrounding trachea after intratracheal injection. Some of the colloids are in lymphatic vessels.

after entry into the lymphatic system, particles probably reside in lymphoid tissue for some time in moving along lymphatic channels to the lymph nodes. In addition to supplying a radiation dose to the peribronchiolar lymphoid tissue, it is also probable that these particles and those in adjacent parenchymal regions are sources of a radiation dose to the bronchiolar epithelium as suggested[62] recently for PuO_2. This aspect of the bronchiolar dose is not often considered. Finally, the accumulation in lymph nodes, the apparently long residence time there and the radio-sensitivity of lymphoid tissue, points to the necessity of considering these organs in an assessment of the radiation hazard.

On the other hand, some aspects of this question lend caution to precipitous selection of lymphoid tissue as a "critical" tissue. The intra-organ distribution of particles is probably such that the most radio-sensitive cells of the organs receive the smallest dose. Furthermore, it is probable that all nodes do not accumulate particles to the same extent.[62] In addition, the reserve capacity of the lymphoid system is apparently large so that one does not know the extent of damage which must be produced before the loss becomes detrimental to the organism. Finally, the accumulation of large masses of material undoubtedly decreases the radiation dose delivered owing to self-absorption. The role of the lymphatic system in an inhalation hazard requires consideration of the factors of distribution, residence time and sensitivity, without which an underestimate of the hazard might occur, and of the facets of intra-organ distribution, physiological reserve and physical radiation absorption, without which considerations one might significantly overestimate the hazard.

4. *Solubilization*

Although we have been considering here "insoluble" particulates, it is axiomatic that no material is totally insoluble. According to the lung model, material considered to be insoluble is not provided for in terms of any amounts of the substance which might be dissolved and absorbed during a long residence time in the lung. It is to be expected that the smaller the particle the greater would be the amount of material solubilized. With material approaching colloidal dimensions, one might expect more rapid and extensive solubilization and systemic absorption even for "insoluble" substances.

Studies with polonium-210 colloid illustrate this point. It was found[14] that with increasing times after inhalation or intratracheal injection, the rosettes of tracks produced by the radiocolloids became progressively less regular in appearance and showed more variance from the initial appearance of a radial orientation. By 30 days after administration, very little of the typical autoradiographic manifestation was found. Rather, one finds clumps and groups of tracks suggestive of radiocolloid which had undergone a "disorientation" or "solubilization".

It is interesting that this disorientation apparently occurred more rapidly with colloids which had been engulfed by macrophages, and was greater as one observed them at higher levels of the respiratory tree. The suggestion has been made[14,46] that material thus delivered to the G.I. tract might have absorption characteristics different from that entering the G.I. tract directly.

Solubilization of the polonium radiocolloid also appeared to be very rapid in lymphoid tissue of the lung. Only rarely were typical autoradiographic manifestations observed and these were in the small masses of lymphoid tissue presumably near the site of entry. In larger lymphoid masses, disorientation was the rule.

It is possible that material entering the lymphatic system in colloidal form might not remain in lymphatic tissue in the same form or for as long a time as might be expected but might contribute to the systemic burden. In using the lung model for material on which insufficient data are available, a consideration of the contribution of solubilization of material, especially with small particles or colloids, is strongly indicated.

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A SIMPLIFIED β-RAY EXTRAPOLATION CHAMBER*

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Abstract—The design and calibration of an extrapolation chamber for the measurement of surface dosages emanating from β-sources is described. The unit can be used to measure dosages from β-ray plaques or other sources of various sizes and intensities. It is a versatile low-cost instrument of quite simple construction. It is designed for direct insertion into the multiple resistor turret of a commercial vibrating-reed electrometer.

THERE are many problems associated with the use of radioactive materials in medical and biological applications which involve the measurement of absolute β-surface doses. In general, the instrumentation for accomplishing this has been complicated and expensive, and therefore has been available to relatively few laboratories.

It can be seen through the work of FAILLA,[1] LOEVINGER[2] and others[3–6] that methods of surface measurement of plane β-sources can be developed on the basis of the cavity-chamber theory. This theory can be applied by utilizing a variable spacing, parallel-plate ionization chamber. The procedure then is to measure ion currents in the ionization chamber for different separations of the parallel plates in the chamber. This paper describes the design and calibration of such an ionization chamber, called an extrapolation chamber, for the measurement of dose rate from beta sources used in the calibration of β-skin dose film packs. This project was a part of the 1957 Nevada Field Test operation.[7]

DESCRIPTION OF THE INSTRUMENT

This instrument was primarily designed to fit directly into the resistor turret of an Applied Physics Corporation vibrating-reed electrometer, which is an effective instrument for measuring small ion currents. The turret contains four Victoreen Hl-Meg resistors (10^8, 10^{10}, 10^{12}, 10^{14} Ω). Calibration of these resistors was made by the Electronics Section and the manufacturer's figures were verified.

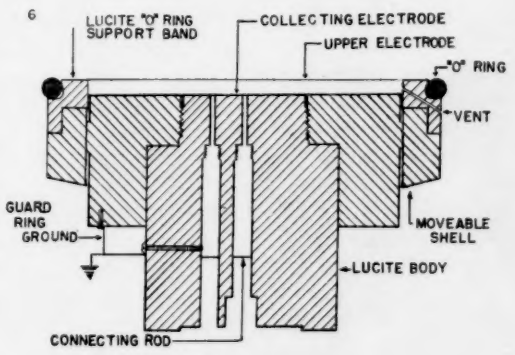

FIG. 1.

A cross-sectional diagram of the extrapolation chamber is shown in Fig. 1. The thin upper electrode is made of 0.00025 in. Mylar film which has been coated on the inside surface with "Neolube" colloidal graphite. Mylar was chosen because of its strength, uniform thickness, and stretch characteristic which allows the electrode to be pulled to tightness after placement of the rubber "O" ring. This last feature is especially desirable as it provides an easily

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obtainable smooth, wrinkle-free surface. The size of any given source to be calibrated is not limited because the instrument can accommodate sources of greater diameter than the head itself. In the cases where a source is not easily moved, the chamber can be faced to it in any position.

A total of six collecting electrodes have been made. Lucite was chosen for its similarity to the scatter characteristics of tissue. The surface was made conductive by evaporating on it a thin film of gold. This very thin gold film has low resistance and because of this thinness, scatter does not become a factor in the measurements. Square separation grooves were cut before the gold coating was applied and it was found that there was no conductivity across these grooves due to the shadow effect in the evaporation process.

The first central electrode (Fig. 2) has three collecting areas of 3, 10 and 30 mm diameter. Two brass screws allow quick change of connections from one electrode size to the next with the nearest ring outside the collecting area being used as a guard ring. The 3 mm electrode makes direct contact with the collecting electrode of the vibrating reed electrometer. When screw *A* has been replaced with a $1\frac{1}{2}$ in. screw, and this is turned in all the way, all three connecting rods are joined so that the collecting area is 30 mm in diameter, and the brass guard ring is in effect. When screws *A* and *B* are in position as shown in Fig. 2, the 3 and 10 mm connecting rods are joined to form a collecting area 10 mm in diameter and the third concentric circle has been added as a guard ring by grounding through screw *B*. When screw *A* is backed out far enough to clear the center rod and grounded, the 3 mm diameter collecting area connects directly to the electrometer and the second and third concentric circles are added to the guard ring. Positive electrical contact is achieved by making the connecting rods to the collecting electrode of spring bronze. This keeps a spring-like pressure on the brass screws at all times; it also serves to keep the metal-to-metal contacts clean.

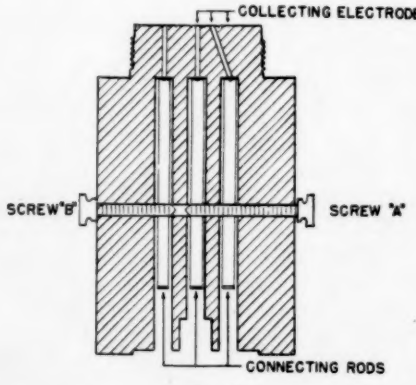

FIG. 2.

Greater electrical stability for sources of lesser strength than can be used with the above (200 mreps/hr for the 30 mm size) is achieved by changeable central electrodes, one of which is shown installed in Fig. 1. Five of these have been made and can be easily interchanged. They have collecting electrode sizes of approximately 1, 3, 8, 16 and 30 mm. The collecting electrode on each has direct shielded connection with the electrometer head providing excellent electrical stability. Each electrode except the 30 mm size uses a common guard ring ground. The 1 mm electrode is a graphite pencil lead which sits on a gold foil on the center rod. The graphite-to-gold contact prevents corrosion and insures electrical contact. All of the grooves have been made 0.005 in. wide. This separation has proved to be sufficient to prevent arcing at the highest voltages applied.

The electrode spacing is controlled by a moveable brass shell threaded and marked externally in micrometer drum style, forty threads per inch and twenty-five divisions around. Each division on the drum represents 0.001 in. vertical separation of the two electrodes. Three small vents drilled in the plastic "O" ring support band admit air and keep the chamber at atmospheric pressure at all separations.

Some trouble with arcing between the upper thin electrode and the collecting electrodes was encountered during the early calibration stages when the thin electrode was accidentally depressed. A 1 MΩ resistor across a neon lamp solved this problem. Shorts in the system do not cause damage after the current has been shunted through the resistor, and the neon indicator gives visual warning.

FIG. 3. Extrapolation chamber (top view).

Fig. 3 shows a top view photograph of the variable size collecting electrodes in the instrument.

CALIBRATION

The original calibration was done with a series of sources including uranium metal, two Tracerlab Sr^{90}–Y^{90} medical applicators, and plane sources used in film dosimetry by this unit. Results agreed with those obtained from an extrapolation chamber borrowed from Atomics International, and in the case of the medical applicators, to within 1 per cent of the Tracerlab calibrations, corrected for decay at the time of measurement. Results were obtained by the use of the following equation:[3]

$$I = 3 \times 10^9 \times \frac{TE}{PRV} \times K$$

where:

I = rate of ionization expressed as e.s.u./cm³ per sec or reps/sec;
P = barometric pressure at time of measurement (mm Hg);
T = absolute temperature at time of measurement;
E = electrometer potential (V);
R = resistance of electrometer circuit (Ω);
K = 760/273;
V = volume of air (cm³) from which ions are collected.

It can be assumed that due to the similarity of the scatter characteristics of lucite and human tissue these measurements represent a close approximation to tissue dose.

Care must be used in the placement of sources on the chamber. Improper alignment can cause air gaps or thin electrode depressions. Compensation can be made for the latter problem by an arbitrary zero point determination. This does not affect the calibration since the slope of the calibration curve is the deciding factor in all cases. Source material must remain undisturbed during an entire series of measurements. Reversal of collecting voltage compensates for the charge carried by β-particles.[3] Under ordinary conditions, reversal is not necessary as the slope remains unchanged, though the zero point differs slightly.

Voltage was varied from source to source, with a minimum voltage being applied to cause saturation without undue thin electrode depression. The Tracerlab medical applicators with their dose rates of 106 and 126 reps/sec needed 150 V to reach saturation. Sources in the mreps/hr range needed only 50 V to reach saturation.

For best results, extrapolations were started with an electrode separation of 0.050 in. This separation was then reduced in steps of 0.001 in. or 0.002 in. Readings at greater separations were beyond the useful portion of the extrapolation curves. Fig. 4 shows a typical curve. The rate of ionization was determined to be 106 e.s.u./cm³ per sec based on the following values taken from this curve and determined by use of the preceding equation. $P = 735.6$ mm Hg; $T = 293°K$; $E = 11.1 \times 10^{-3}$ V; $R = 8.4 \times 10^7\ \Omega$; $V = 4.1 \times 10^{-3}$ cm³.

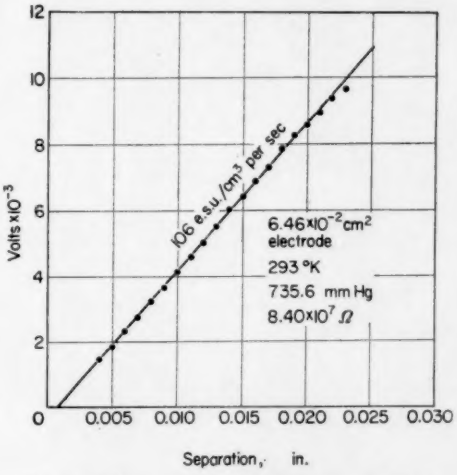

FIG. 4. Measurement of strontium applicator.

All sources were finally extrapolated through a total of 6.8 mg/cm² of Mylar film, including the thin electrode. This was adequate to stop the α-particles in the uranium and simulate a skin exposure condition for all the sources. Simulated tissue depth–dose curves were also obtained by using tissue equivalent absorbers between the source and the thin electrode.

SUMMARY

This paper has shown a method used at this laboratory for the measurement of beta

radiation through the use of a vanishing chamber type extrapolation instrument.

A small, versatile and inexpensive chamber was designed, constructed, and calibrated for use in conjunction with an Applied Physics Corporation vibrating-reed electrometer. Six interchangeable central electrodes of assorted diameters accommodate sources of a variety of sizes.

β-calibrations were made and the unit agreed within 2 per cent with the standardized sources, namely natural uranium metal placques, and two Tracerlab Sr^{90}–Y^{90} medical applicators.

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STRUCTURAL IMPLICATIONS IN CESIUM SORPTION

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Abstract—The results of this study help to explain the effectiveness of the local Conasauga shale for removing cesium from radioactive waste solutions. Illite, which is present in the shale has a particularly high affinity for cesium. The importance of the *c*-axis dimension of the 2:1 layer lattice clays was demonstrated by using synthetic micas and selected cation treatment. The beneficial role of potassium in the mica and illite is primarily because this element can induce and maintain collapse of the *c*-axis. Ion exchange capacities and surface area are secondary to the *c*-axis dimension in predicting the cesium sorption behavior of clays.

The potassium treatment and oven-drying of clays followed by cesium sorption may be used to differentiate montmorillonite from vermiculite; the importance of the findings in terms of weathering and diagenetic processes involving clays is discussed.

INTRODUCTION

IRRESPECTIVE of the ultimate mode of disposing of radioactive fission product wastes, knowledge of the possible interactions between the fission products and soils is of prime importance. Sorption reactions are of particular interest since several disposal schemes in operation utilize the ion exchange capacity of pure clay minerals and of soils to decontaminate waste solutions. At Oak Ridge National Laboratory, for example, a conventional lime-soda softening plant is used to process low-level waste solutions; this type of treatment is effective in reducing the radiostrontium concentration of the waste solution. Addition of the clay mineral illite during the "softening" operation was found to be effective in reducing the radiocesium concentration.[1]

In the disposal of intermediate-level wastes, the supernatant liquid, resulting from the neutralization of the waste in storage tanks, is pumped to pits excavated in the local Conasauga shale formation.[2] About 75 per cent of the activity in the supernate is due to radiocesium; seepage of the solution through the shale formation results in a high degree of removal of this element from the waste. Sorption is by far the most important reaction involved in the removal. Although high-level wastes are now stored in tanks, potential methods of disposal include injection of these wastes into deep geologic formations. In these studies, too, sorption plays an important role, since preferential sorption and consequent concentration of radionuclides by clay minerals may result in excessive heat generation.

Numerous other examples could be cited to show the implications of sorption reactions; however, the above examples are sufficient to justify investigation of the mechanisms involved in cesium sorption. This paper reports the results of tests made with several clay minerals, including the local Conasauga shale, and discusses sorption of cesium by considering the effect of the *c*-axis dimensions of the clay minerals.

DESCRIPTION OF SAMPLES

The illite (Fithian, Ill.), montmorillonite (Clay Spur, Wy.) and kaolinite (Macon, Ga.) samples were obtained from Ward's Natural Science Establishment. The vermiculite sample is from Travelers Rest, South Carolina, and

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

was donated by the Zonolite Co. X-ray diffraction analysis showed that this material called vermiculite is regularly interstratified biotite and vermiculite with minor amounts of hornblende. Mineral allocations based on X-ray diffraction, chemical, base exchange and heating-weight-loss data gave a vermiculite concentration of 45 per cent. Although an estimate of the error is difficult, an approximate error of ± 10 per cent is probable. Since the material is a 1:1 interstratified system, approximately 45 per cent is biotite; the remainder is mostly hornblende.

Studies made by the U.S. Geological Survey show that hydrous micas and kaolinite are the principal clay minerals in the Conasauga shale. The term *hydrous mica* does not necessarily define any clay mineral; in fact, it implies interstratification of mica (illite*) with an expanding lattice mineral, such as montmorillonite or vermiculite. The material used in this study was examined by X-ray diffraction methods. From the diffraction diagrams, total chemical analysis and base exchange data, the approximate mineral composition of the shale was determined. The principal clay mineral was illite (40 per cent); also present in appreciable quantities was a mineral showing a diffraction maxima at 14 Å (25 per cent). Although the 14 Å maximum could be due to either vermiculite, chlorite or montmorillonite, later studies (discussed in a separate section of this paper) strongly supported the mineral in question to be vermiculite. The remaining 35 per cent was quartz.

DESCRIPTION OF THE SYSTEM

Waste solutions do not contain radioactive elements as the major ionic constituent. Depending on the source of the waste solution the major constituent could be H^+ ions (Purex waste and most laboratory waste), Na^+ ions (neutralized Purex) or Al^{+++} ions (acid aluminum nitrate waste). For most of the studies conducted in this series of experiments, the Al^{+++} concentration was 1.67 M when simulated $Al(NO_3)_3$ waste was used, and the Na^+ concentration 6.0 M when neutralized Purex concentrate was being simulated. The cesium concentration was limited to the range of $0.2–8.0 \times 10^{-4}$ moles/l.; this level of cesium concentration would be found in high-level wastes.

For analytical purposes Cs^{137} was added; cesium sorption was then measured by counting the waste solution before and after being in contact with the clay mineral. A 1-ml aliquot was counted in a well-type scintillation counter with a NaI(Tl) crystal. Solution-clay mineral contact time was limited to 16 hr with the exception of studies for which time was considered a factor in sorption.

SORPTION OF CESIUM

The ability of the Conasauga shale to remove radioactive fission products from waste solutions has been reported previously.[4,5] The interesting feature of this removal is the strong preference exhibited by the shale for cesium. In Fig. 1 the sorption of cesium from 1.67 M $Al(NO_3)_3$ solution is shown for the local shale and several clays. Of the materials tested, shale has the highest affinity for cesium; furthermore, with the mineralogical composition known for the shale, the high capacity for cesium by illite is suggested from the data in Fig 1. Tests performed on illite, which was not available during the early stages of experimentation, confirmed this belief (Fig. 2).

Several other interesting features are evident from the figures. In the lower cesium concentration range (high Al/Cs ratio), illitic shale sorbed the highest amount of cesium; vermiculite is also effective. With increasing cesium concentration, montmorillonite surpasses vermiculite for sorbing cesium. One of the surprising features is the high specificity exhibited by illite, a mineral with only a moderate ion exchange capacity when compared with montmorillonite or vermiculite.

Changing the system to a 6.0 M $NaNO_3$ waste type did not affect the sorption of cesium to any great degree (Fig. 3). Note again that illite is more effective than montmorillonite on a

* The nature and relationship of illite to mica are being investigated by H. S. Yoder and his colleagues of the Carnegie Institution of Washington; the reader is referred to their paper for a discussion of the subject and for further references.[3]

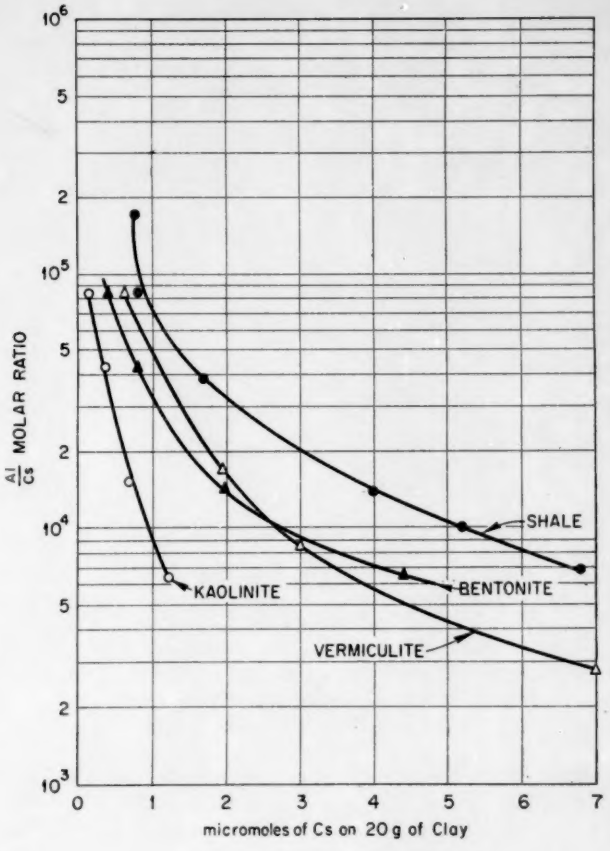

FIG. 1. The sorption of cesium by clays and shale from 50 ml of 1.67 M aluminum nitrate waste solution and 20 g of material.

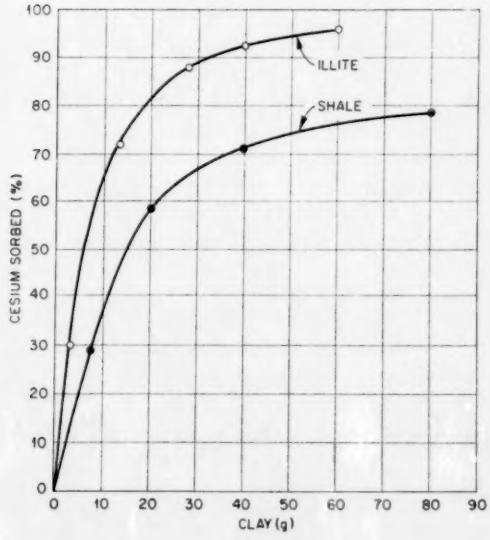

FIG. 2. Cesium sorption by illite and Conasauga shale; 50 ml of 1.67 M aluminum nitrate solution containing 50 mg/l. of cesium was used.

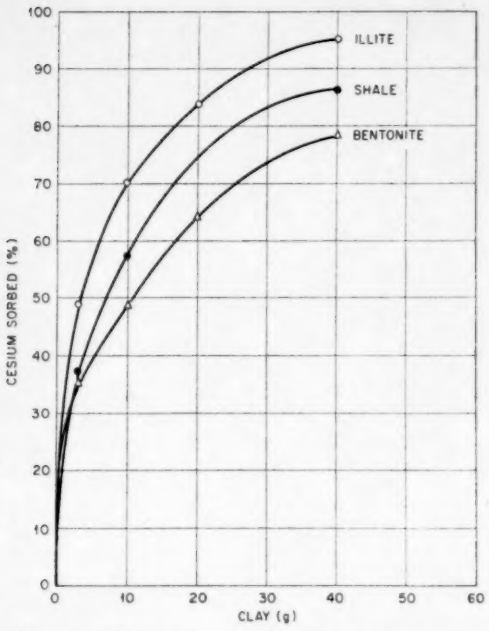

FIG. 3. Cesium sorption by illite, montmorillonite (Wyoming bentonite) and Conasauga shale; 50 ml of 6.0 M sodium nitrate waste containing 50 mg/l. of cesium was used.

weight (or exchange capacity) basis. In Table 1 the sorptive capacity of the various materials for cesium is shown; the results are expressed on the basis of both weight and exchange capacity. On either basis illite sorbs the highest amount of cesium in the concentration range employed. The results also strongly suggest that sorption by illite is dependent on contact time; sorption by Conasauga shale also shows some evidence of time dependence, which would be expected since illite is the dominant clay mineral.

Although the effectiveness of kaolinite based on weight is low, it is quite effective on the basis of its ion exchange capacity. Both montmorillonite and vermiculite are not particularly effective based on their ion exchange capacity.

CESIUM SORPTION BY SYNTHETIC MICAS

The results of these preliminary investigations show that illite is an outstanding clay for sorbing cesium from both aluminum nitrate and sodium nitrate waste solutions. If ion exchange capacity *per se* cannot explain the differences in the clays' ability to sorb cesium, one must look to other distinguishing features to explain the observed behavior of the clay minerals. There are two immediately apparent features by which illite differs from montmorillonite and vermiculite. Illite has a *c*-axis spacing of 10 Å, whereas montmorillonite and vermiculite have spacings greater than 10 Å which depend on the degree of hydration. Illite also contains a high amount of potassium in its chemical make-up, and these potassium ions are accommodated between the aluminosilicate sheets.

To test the effect of the *c*-axis spacing on the sorption of cesium, two synthetic mica minerals were used.* One of the micas was a potassium fluorophlogopite which is very similar to illite. The other mica, sodium fluorophlogopite, differs from illite by having sodium ions in the positions normally occupied by potassium ions. Both minerals have the layer-lattice structure and exhibit 10 Å spacings along the *c*-axis. The results of the sorption tests are shown in Fig. 4. The curve obtained by illite is also included for comparison.

The results support the thesis that cesium sorption is favored by the 10 Å spacing. The higher sorption of cesium by sodium fluorophlogopite suggests that the beneficial role of the potassium ion in illite is in inducing and maintaining the collapsed state rather than any special influence on ion exchange capacity. Ion exchange determinations gave the following results for particles <140 mesh: Illite = 15.4 mequiv/100 g, K-fluorophlogopite = 13.8 mequiv/100 g and Na-fluorophlogopite = 5.6 mequiv/100 g. Note that although Na-fluorophlogopite has the lowest ion exchange capacity, it is the most effective for the sorption of cesium.

CESIUM SORPTION BY POTASSIUM-SATURATED CLAYS

The evidence presented so far emphasizes the importance of the *c*-axis dimensions for the sorption of cesium. If the 10 Å spacing of the micas and illite is desirable for sorption, then

* The synthetic micas were furnished by the Bureau of Mines' Electrotechnical Laboratory in Norris, Tennessee.

Table 1. *Sorption of cesium by clays from* 6 M $NaNO_3$ *waste solutions*

Clay	Cation exchange capacity (mequiv/g)*	Weight of cesium sorbed			
		mg/g* of clay		mg/mequiv of exchange	
		Contact time			
		18 hr	90 hr	18 hr	90 hr
Illite	0.15	0.164	0.170	1.09	1.13
		0.139	0.151	0.93	1.01
		0.143	0.156	0.95	1.04
		0.152	0.184	1.01	1.23
Montmorillonite	1.00	0.157	0.140	0.157	0.140
		0.142	0.142	0.142	0.142
		0.141	0.141	0.141	0.141
Vermiculite†	0.57 (1.27)	0.052 (0.091)	0.051 (0.089)	0.041 (0.072)	0.040 (0.046)
		0.039 (0.068)	0.034 (0.060)	0.031 (0.054)	0.026 (0.031)
		0.040 (0.070)	0.043 (0.075)	0.032 (0.056)	0.034 (0.060)
Kaolinite	0.087	0.036	0.026	0.414	0.299
		0.040	0.032	0.460	0.368
Conasauga shale	0.27	0.104	0.107	0.385	0.396
		0.086	0.098	0.318	0.363
		0.084	0.086	0.311	0.318

* All values based on the oven-dried weight of the clay.

† Values in parentheses refer to calculated sorption of the pure mineral.

conversion of the higher spacings of montmorillonite and vermiculite to 10 Å should improve their sorption characteristics. One of the methods used to induce collapse is to saturate the clays with potassium ions; vermiculites show a 10 Å spacing, and montmorillonites show an 11.9 Å spacing following potassium saturation.

In Table 2 the results of sorption are given for the various materials, including the local Conasauga shale. Illite was only slightly affected by the potassium treatment; this is to be expected since illite contains potassium initially. Vermiculite and Conasauga shale show the highest amount of change in their abilities to sorb cesium. The spacing for the *c*-axis of vermiculite was found to be reduced from 14 to 10 Å. Montmorillonite does show an increase in its ability to sorb cesium; the *c*-axis spacing was reduced from 12.6 Å in the air-dry state to 11.9 Å.

Sorption tests were also made on the potassium-saturated clay samples after they were dried in the oven at 100°C. The vermiculite and Conasauga shale samples showed an even greater improvement in their abilities to sorb cesium. However, oven drying affected neither the montmorillonite nor the kaolinite samples. Structurally, the materials differed only in degree from the potassium-treated air-dried samples; the vermiculite samples showed a sharp 10.4 Å spacing in contrast to a peak which was broadened towards the high-spacing side when air-dried. The

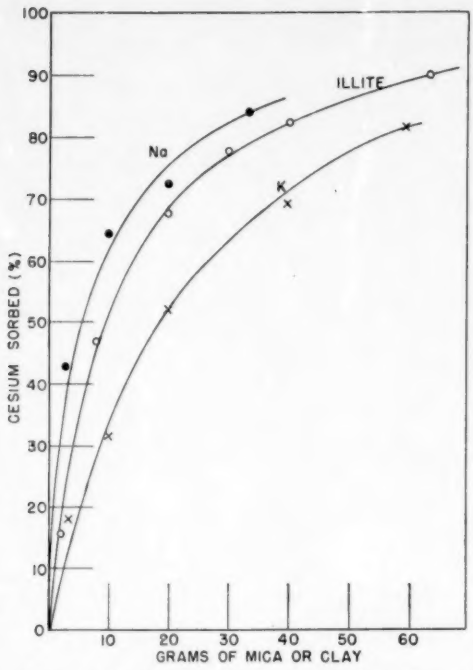

FIG. 4. Cesium sorption by synthetic sodium and potassium fluorophlogopites; 50 ml of 6.0 M sodium nitrate waste containing 100 mg/l. of cesium.

montmorillonite sample showed almost identical spacing and peak characteristic for both air- and oven-dried samples.

Samples which were oven-dried but not potassium-saturated did not show an increase in sorption for cesium. This behavior is in agreement with the concept that the water associated with the clays is related to the cation on the clays.[6] It is also interesting to note that cesium sorption is a function of time, particularly when potassium is the saturating cation. Here again, the important parameter affecting sorption as a function of time is the *c*-axis spacing. This belief is supported by the data obtained from the synthetic micas. With the sodium fluorophlogopite, the sorption of cesium was as follows: 14.1 per cent sorption after 1 hr; 45.9 per cent after 18 hr; and 68.2 per cent after 90 hr.

DISCUSSION

The results of the experiments show that ion exchange capacity alone will not explain the quantitative sorption of cesium. One can extend these results on the basis of known surface area of clays, and state that surface area alone will not explain sorption by clays.

From these results, it is evident why the local shale is effective in reducing the concentration of cesium in the waste solution seeping through the formation. The high content of illite in the Conasauga shale is responsible for the decontamination. Furthermore, following the identification of illite in Conasauga shale, experiments were conducted using grundite (commercially available illite) in the waste water treatment plant operating at Oak Ridge National Laboratory for the removal of cesium. The results show that about 85 per cent of the cesium is removed by 150 p.p.m. of the clay.[1] The recommendation to use grundite was made prior to the full realization of the role of the lattice parameter in cesium sorption; from the results shown in Table 2, it would be interesting to see the effectiveness of potassium-saturated vermiculite and Conasauga shale in the removal of cesium in the waste water treatment plant.

Apart from the direct application of the results to the study of radioactive waste disposal, several implications of the results become apparent. A particularly interesting sequence is the sorption of cesium by vermiculite. In the natural state it shows a definite affinity for cesium; a discrimination factor* of about 7 was calculated. Following potassium saturation, a discrimination factor of 28 was calculated based on the exchange capacity of the natural material; if the exchange capacity of the potassium-saturated material is used (16 mequiv/100 g) to calculate the discrimination factor, a value of 100 is obtained. An analogy of the behavior can be made with the vermiculite–potassium system since both potassium and cesium are known to behave similarly in inducing collapse of the *c*-axis. The potassium–clay relationship represents one of the most significant systems in agriculture and mineralogy. In the weathering of micas,

* The discrimination factor is derived by dividing the Na/Cs ratio in the original waste solution by the Na/Cs ratio of the clay.

Table 2. Sorption of cesium by clays from 6 M $NaNO_3$ *waste solutions after potassium saturation*

Clay	Weight of cesium sorbed (mg/g*)			
	Air-dried samples		Oven-dried samples	
	Contact time			
	18 hr	90 hr	18 hr	90 hr
Illite	0.142 0.120 0.126	0.142 0.118 0.132	0.139 0.138	0.145 0.141
Montmorillonite	0.172 0.172 0.173	0.188 0.179 0.178	0.173 0.169	0.176 0.170
Vermiculite†	0.090 (0.158) 0.088 (0.154) 0.065 (0.114)	0.097 (0.170) 0.096 (0.168) 0.076 (0.133)	0.106 (0.186) 0.088 (0.154) 0.098 (0.172)	0.120 (0.210) 0.113 (0.198) 0.122 (0.214)
Kaolinite	0.060 0.050	0.041 0.041	0.031 0.024	0.038 0.016
Conasauga shale	0.150 0.142 0.128	0.143 0.139 0.129	0.227 0.280	0.200 0.301

* All values based on the oven-dried weight of the clay.
† Values in parentheses refer to calculated sorption by the pure mineral.

JACKSON *et al.*[7] consider the driving forces for removing potassium to be water and cations, and that the depletion of potassium along a given plane is enhanced by a weathering rate function. The results obtained from the cesium sorption studies on vermiculites suggest that the depletion along a given plane is due to reduced affinity for potassium by the increase in spacing along the plane of entrance of water.

Another interesting consideration from these results is the increasing evidence that clay minerals occur as interstratified systems. One of the most frequent associations is the vermiculite– and montmorillonite–mica systems.[8] Since the evidence on cesium sorption reveals that the affinity for cesium (and therefore potassium) is increased when the interlayer spacing is reduced, this would offer an explanation of how layers can be completely potassium-saturated and collapsed leaving other planes relatively unsaturated with potassium, and in an expanded state. These studies are of value then to anyone interested in weathering and diagenesis of clays.

The behavior of the clays upon potassium saturation and oven-drying suggests the use of cesium sorption as another aid in the identification of clay minerals. Thus, high cesium sorption by a relatively low base-exchange clay suggests illite; potassium saturation of the system should aid in telling one whether montmorillonite and/or vermiculite is present. Oven-drying of the sample followed by cesium sorption should help in differentiating montmorillonite from vermiculite. Thus with the Conasauga shale, X-ray diffraction patterns were not conclusive, since the material showed rather poorly defined diffraction maxima. One

of the evidences which favored the assignment of the 25 per cent mineral component to vermiculite was the observed behavior of the shale after oven-drying the potassium-saturated clay. If montmorillonite were present, it would be highly unlikely that oven-drying would have caused an increase in cesium sorption as was evidenced by the shale.

Acknowledgment—The authors wish to acknowledge the encouragement and assistance of E. G. STRUXNESS in the course of this study.

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A HIGH-ENERGY NEUTRON-FLUX DETECTOR*

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Abstract—High-energy neutrons and protons in the range 20.4 MeV to more than 400 MeV can be detected in 1720-g plastic scintillators by means of $(n, 2n)$ and (p, pn) reactions with carbon nuclei. Carbon-11 is formed which emits positrons of $E_{max} = 0.98$ MeV and has a half life of 20.4 min. With scintillators irradiated to saturation in a neutron flux density of 1 neutron cm^{-2} sec^{-1}, a subsequent 5-min waiting period and a 38-min counting time, the standard deviation in the counting statistics is less than 15 per cent. Background considerations are discussed along with some experimental and theoretical values of the $(n, 2n)$ reaction cross sections.

I. INTRODUCTION

A METHOD of detecting both neutrons and protons covers the energy region from 20.4 MeV[1] to more than 400 MeV. It involves the production of carbon-11 from carbon-12 by $(n, 2n)$ and (p, pn) reactions with carbon nuclei. Carbon-11 is a positron emitter of $E_{max} = 0.98$ MeV. The half life is 20.4 min, and there are no significant competing reactions. The $(n, 2n)$ reaction cross sections are fairly constant above about 50 MeV.

Carbon in the form of plastic scintillator material is used as the detector. These scintillators, 5 in. in diameter and 5 in. high, weigh 1720 g each. Not only is the energy of the positron expended entirely in the scintillator, but the energy of one or both of the annihilation gammas is likewise largely contained.

II. COUNTING SYSTEM

The scintillators are exposed in an area to be monitored for a suitable time (preferably to saturation for low flux densities), and are then counted in an area of lower background. As with other threshold detectors, this method is insensitive to accelerator-produced gamma rays.

These scintillators are counted in a mineral-oil bath on top of a vertically mounted Du Mont 6364 photomultiplier. The light-tight housing for the photomultiplier is a mild steel box 20 in. high, 8 in. on each side, and 1/4 in. thick (see Fig. 1). Mild steel was chosen because of its value as a magnetic shield, since on occasion C^{11} has been counted near the Bevatron. In addition to the lid on top, there is another opening on the side, near the bottom, for access to the phototube base.

A white paint, Kerpro No. 22 (titanium dioxide pigment in a clay base), was used to spray all but the viewing end of each scintillator. Fig. 2 shows how painting the scintillator increased the gain, as indicated by the lower setting of the high voltage. The curves are differential pulse-height distributions from a Co^{60} source.

Pulses from the photomultiplier are fed to a preamplifier, after which they are clipped so that all pulses have a uniform width of 2 μsec. The pulses are then fed into a UCRL Model 5 linear amplifier, from there to an Atomic single-channel pulse-height analyzer, and then to a scaler (see Fig. 3). The resolving time of the entire system is less than 5 μsec. It is necessary to gate the scalers off during the acceleration phases of the Bevatron and 184-in. cyclotron cycles, even though the counting equipment is normally some distance away.

The Bevatron is gated off by a telemetering unit which produces a gating pulse at the start of the acceleration cycle. This pulse, fed into a

* Work done under the auspices of the U.S. Atomic Energy Commission.

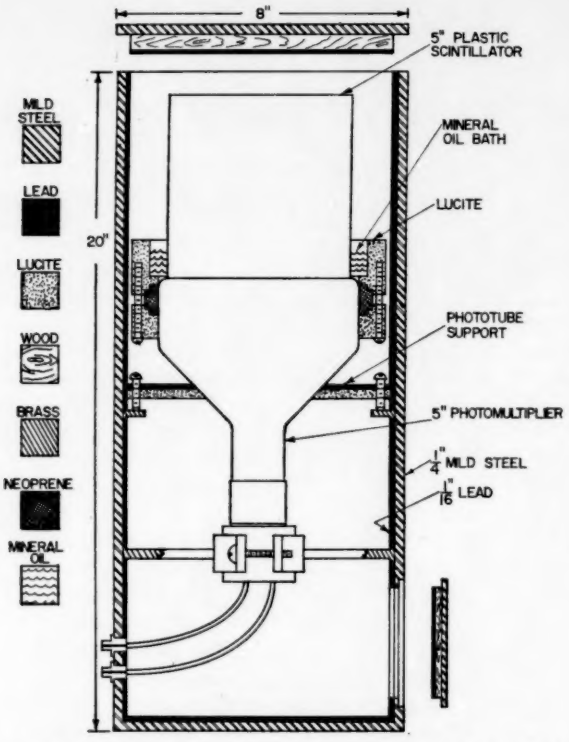

FIG. 1. Photomultiplier housing assembly, showing 1/4-in. mild steel sides, 1/16-in. lead liner, vertically mounted DuMont 6364 photo-multiplier, and plastic scintillator immersed in mineral oil bath.

variable gate and delay chassis, is shaped and stretched to form a 3-sec-long off gate. A 1000-cycle pulse-generator signal is fed into a similarly gated scaler in order to keep track of counting time.

A cyclotron triggering signal is run directly into a variable gate and delay chassis and is treated in a similar manner except that the off gates are only a few milliseconds long. However, the repetition rate is high, about 63 pulses/sec.

The loss of counting time due to gating is considerable and is a matter of great concern. The data treatment presented here presupposes no losses of this kind. Often something less than this ideal is realized in practice.

III. BACKGROUND

The counting equipment is in a building with 1-ft-thick heavy aggregate concrete walls. The photomultiplier housing is surrounded by 4 in. of lead. Photomultiplier noise, measured with scintillator removed, did not add appreciably

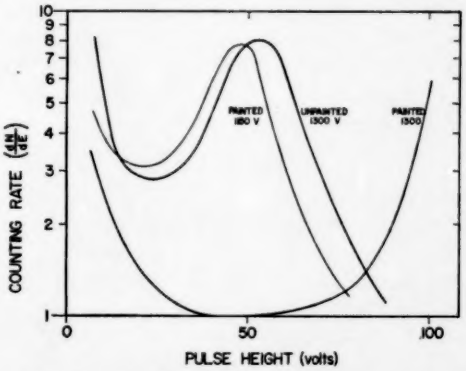

FIG. 2. The effective increase in gain provided by spraying the nonviewing surfaces of the scintillators with a white paint is clearly indicated. A Co^{60} source was used for obtaining the curves.

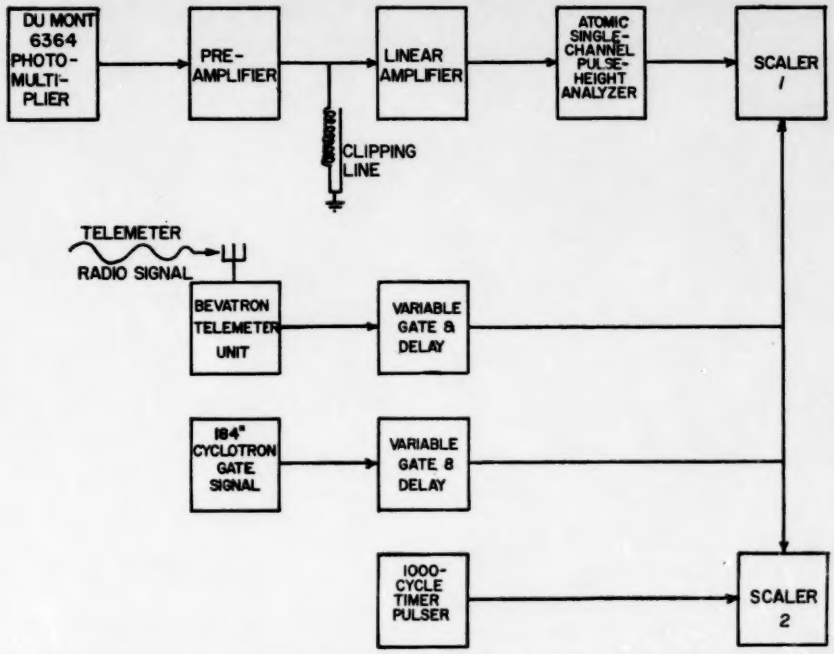

FIG. 3. Carbon-11 pulses from the photomultiplier are clipped, amplified, pulse-height analysed and counted in scaler No. 1 unless scaler No. 1 is gated off (as during the acceleration phases of the Bevatron and 184-in. cyclotron cycles). Scaler No. 2 is similarly gated and counts pulses from an accurate 1000-cycle generator, and this is a measure of counting time.

to the background, therefore it was not considered necessary to view both ends of the scintillator with two phototubes in coincidence.

The 1/16 in. of lead used to line the photomultiplier housing reduced the background (especially in the low-level channels) by a factor of 0.56 between 5 and 10 V, and 0.91 between 20 and 25 V.

At first we expected that background might consist not only of cosmic rays but also of high-energy gamma rays produced by thermal-neutron capture in either the lead shielding or the iron wall. This was shown to be unimportant at present background levels: stacking boron-loaded paraffin blocks around the photomultiplier house did not lower the background counting rate.

We also tried a cosmic-ray anticoincidence shield consisting of plastic scintillator 1 in. thick and 13 in. in diameter, placed over the photomultiplier housing and viewed by two photomultipliers. Background was reduced, but only by 10 per cent which at this stage of development did not seem to warrant the time that would be necessary to maintain properly the anticoincidence system. Certainly, some of the low-activity construction materials, mercury shielding, quartz window phototubes etc. would have done much to reduce background further. Fig. 4, curve C, shows the present background.

A system similar to that used for counting C^{11} has been used simultaneously with the C^{11} system in order to check background during counting of the activated scintillator.

IV. COUNTING CONSIDERATIONS AND STATISTICS

Because accelerator-produced gamma rays are not counted, it is much easier to extrapolate the integral bias curve because there is no confusion between these gammas and small C^{11}-produced pulses near the zero-bias intercept. The shape of the integral bias curves, determined from high-level activations, provided an

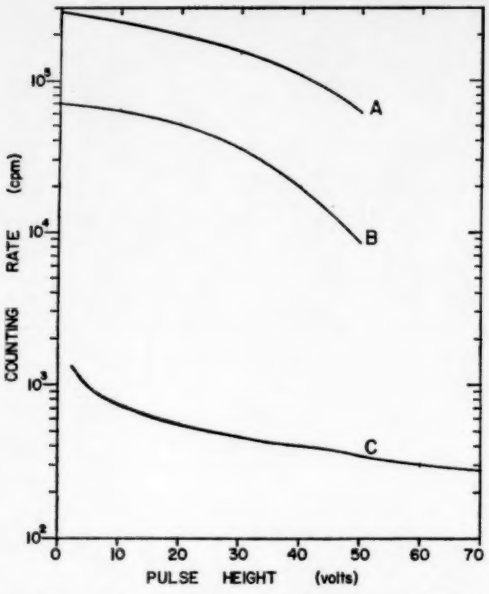

FIG. 4. Curve C is the background integral with 4 in. of lead and without the anticoincidence shield. Integral bias curves A and B obtained at different beam intensities with two different scintillators indicate that no large errors are introduced by making the extrapolation from the 10-V operating point. Also, if many counts were being lost the curves would be more steep in this region. The similarity of the shapes of the curves indicates that corrections for individual scintillators are not necessary. For the two curves shown, the zero-bias correction factors are within 5% at 10 V.

accurate correction factor for the extrapolation to zero bias. Pulses were taken from 5 to 45 V in 5-V steps and then corrected for saturation, decay and background. It is seen in Fig. 4, curves A and B, that the shapes of the integral bias curves obtained from two different scintillators under different beam intensities and corrected for decay become rather flat as the zero-bias intercept is approached. If many counts were being lost the curves would be steeper in this region. Normally, it should not be necessary to make corrections for individual scintillators. For the two curves shown, the zero-bias correction factors are within 5 per cent at 10 V. Integral counts are taken at the 10-V point, since from examination of the C^{11} and background curves the ratio of the standard deviation of the total count to the C^{11} count is a minimum at this point.

Calculation of induced activity

At saturation we have

$$\frac{dN}{dt} = nv\sigma_{(n,2n)}N_t = -\frac{dN'}{dt} \qquad (1)$$

where $\frac{dN}{dt}$ = C^{11} production rate, nv = neutron flux density, $\sigma_{(n,2n)}$ = neutron absorption cross section leading to the production of positron activity, N_t = total number of C^{12} atoms = 7.88×10^{25} atoms C^{12} per scintillator, and $\frac{dN'}{dt}$ = zero-time disintegration rate.

If an incident neutron flux density of 1 neutron cm^{-2} sec^{-1} and a cross section of 22×10^{-27} cm^2 is assumed, we find

$$\frac{dN'}{dt} = 104 \text{ disintegrations/min}$$

After a 5-min delay prior to counting there would be

$$\frac{dN'}{dt} \exp\,(-t_w/T) = 87.7 \text{ dis/min}$$

where T = mean life of C^{11},

t_w = delay time prior to counting.

Of these, 92 per cent possess sufficient height to overcome the lower-level bias and be counted.

The counting errors were computed as follows:

$$N_d = C\int_{t_w}^{t_w+t_c} (dN/dt)\,dt$$

$$= C\int_{t_w}^{t_w+t_c} R_0 \exp\,(-t/T)\,dt \qquad (2)$$

$$= R_0 TC\,\{\exp\,(-t_w/T) - \exp\,[-(t_w + t_c)/T]\} \qquad (3)$$

where N_d = number of disintegrations occurring during the counting period after secular equilibrium has been reached, R_0 = initial decay rate of the scintillator upon removal from the neutron flux, T = mean life, C = zero-bias correction factor, t_w = initial decay time prior to counting, and t_c = counting time.

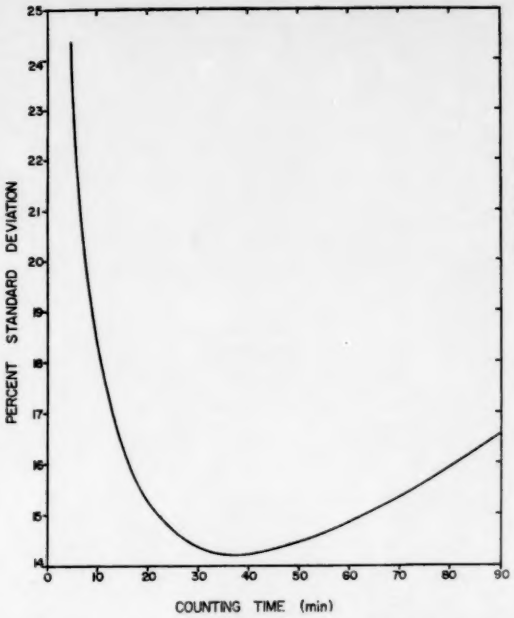

FIG. 5. A special case of equation (4) showing the error associated with the counting statistics as a function of the counting time for an incident flux density of 1 neutron cm^{-2} sec^{-1} for a scintillator irradiated to saturation, and including a 5-min decay period prior to counting. For an optimum counting time of 38 min the standard deviation is less than 15%.

It is assumed that no time is lost by gating, and that the scintillator did not undergo changes in the incident flux level during irradiation. Then,

per cent standard deviation

$$= \frac{100\sqrt{N_d + 2Bt_c}}{N_d} = \sigma \qquad (4)$$

where B is the background (in counts/min). The optimum counting times for minimum standard deviations may be computed by differentiating equation (4) and finding, for various values of R_0, the value of t_c that satisfies the equation $d\sigma/dt_c = 0$,

$$0 = 1 - \exp(t_c/T) + \frac{K}{2BT}\exp(-t_w/T) + \left(\frac{2t_c}{T}\right) - \frac{K}{2TB}\exp[-(t_w + t_c)/T] \qquad (5)$$

where $K = R_0TC$.

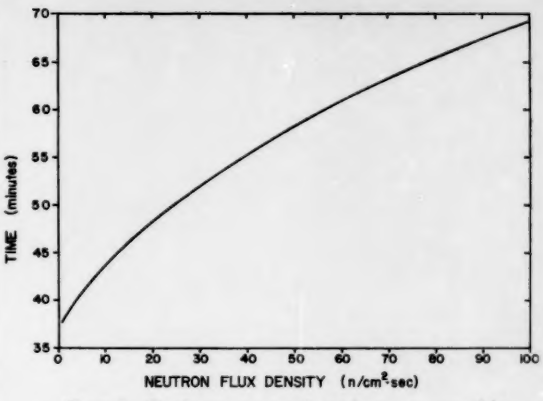

FIG. 6. Optimum counting times to provide minimum standard errors were calculated from equation (5) and are plotted as a function of the neutron flux density.

Fig. 5 is a special case of equation (4) and shows the error associated with the count as a function of the counting time for an incident flux density of 1 neutron cm^{-2} sec^{-1} for a scintillator irradiated to saturation, and including a 5-min decay period prior to counting. Five min or less is usually sufficient time for transporting the scintillator to the counting area. For an optimum counting time of 38 min, this neutron flux density can be determined with a standard deviation of 14.5 per cent. Optimum counting times for other neutron flux densities were calculated from equation (5) and shown in Fig. 6. These values of time and flux density were then substituted in equation (4) and the corresponding values of per cent standard deviation were plotted versus neutron flux density in Fig. 7. Thus a flux density of 20 neutrons cm^{-2} sec^{-1} can be determined with an error of less than 1 per cent when the counting time is 48 min. These figures are standard deviations based on counting statistics and do not include systematic errors or errors in the determination of the reaction cross section.

An upper-level discriminator set at 1.5 or 2 MeV[2] might be an interesting method of further increasing the sensitivity of the system. In effect, then, one would have a single-channel differential pulse-height analyzer with a very large window. This would take advantage of the fact that the integral bias curve for C^{11} falls

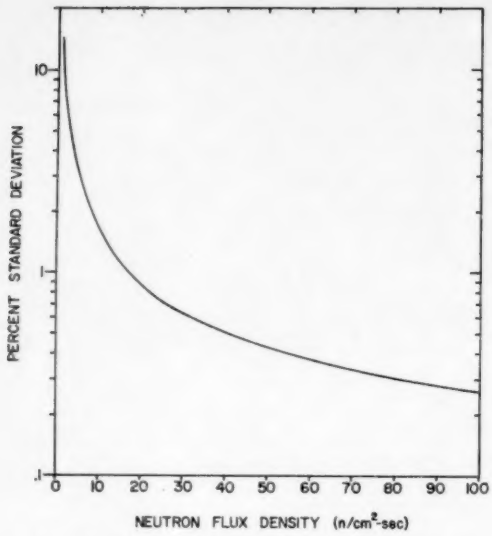

FIG. 7. The optimum counting times shown in Fig. 6 were substituted together with their associated flux densities, in equation (4), and their corresponding standard deviations plotted here. Thus a neutron flux density of 20 neutrons cm^{-2} sec^{-1} can be determined with a counting error less than 1% when a counting time of 48 min is used.

off rapidly at these energies, while the background curve is still relatively flat. Fig. 8 shows the minimum per cent standard deviations as a function of window width at various bias levels for these differential counts. A neutron flux density of 1 neutron cm^{-2} sec^{-1}, secular equilibrium, a 5-min decay and a 40-min counting time were assumed. Thus, for a window looking between 20 and 50 V (a window width of 30 V, and a 20-V bias), a minimum standard deviation of 13 per cent may be realized.

The graph shown in Fig. 9 is a plot of per cent saturation vs. irradiation time,

$$\text{per cent saturation} = 100[1 - \exp(-t/T)] \quad (6)$$

where t = irradiation time, which may facilitate calculations involving irradiations other than those producing saturated activity. An uninterrupted uniform irradiation rate over the entire exposure period is assumed.

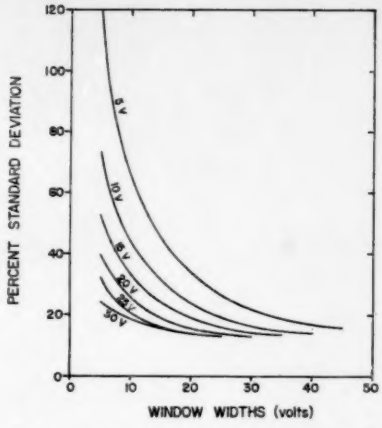

FIG. 8. Effective increase in sensitivity is obtained by adding an upper-level variable discriminator, thus taking advantage of the fact that the C^{11} integral-bias curve falls off rapidly above about 25 V while the background is still relatively flat above this region (Fig. 4). With a bias of 20 V and an upper-level discriminator of 50V (window width = 30 V), the minimum standard deviation for 1 neutron cm^{-2} sec^{-1} is 13%.

In this case we have

$$Nd = KR_0TC\left[\exp\left(-\frac{t_w}{T}\right) - \exp -\left(\frac{t_w + t_c}{T}\right)\right] \quad (7)$$

where K = per cent saturation/100.

V. REACTION CROSS SECTIONS

The cross section as a function of energy of the $C^{12}(p, pn)C^{11}$ reaction is well known[3] (see Fig. 10). Data on the $C^{12}(n, 2n)C^{11}$ reaction cross section have been determined for only a few energies; however, at high energies the cross section should be proportional to the nucleon–nucleon cross section. Based on the data in Figs. 11 and 12 for neutron–neutron and proton–neutron cross sections at high energies,[4] an approximation of the $(n, 2n)$ cross section has been made.[5] We have

$$\frac{\sigma(n, 2n)}{\sigma(p, pn)} \propto \frac{N\sigma(n, n)}{N\sigma(p, n)} = \frac{6(24)}{6(36)} = \frac{2}{3} \text{ at 300 MeV,}$$

where N is the number of neutrons in the carbon nucleus. This ratio, determined for several

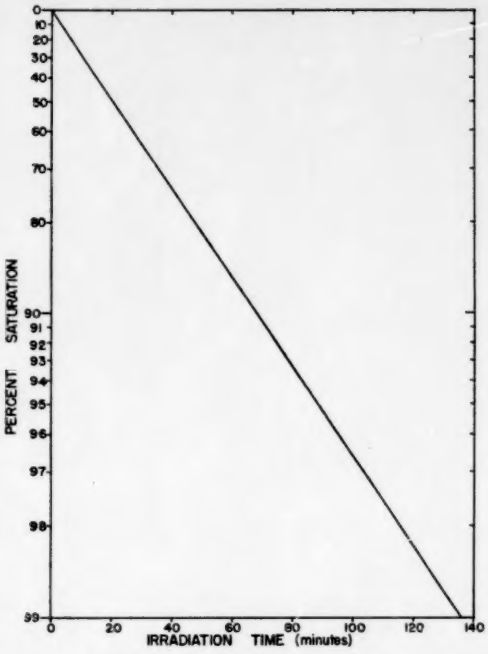

FIG. 9. Per cent saturation shown as a function of irradiation time (to facilitate calculations involving irradiations other than those producing saturated activity).

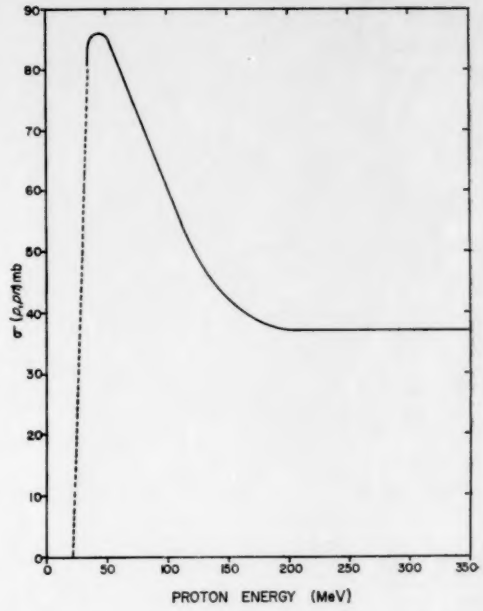

FIG. 10. $C^{12}(p, pn)C^{11}$ reaction cross sections taken from CRANDALL *et al.*[3]

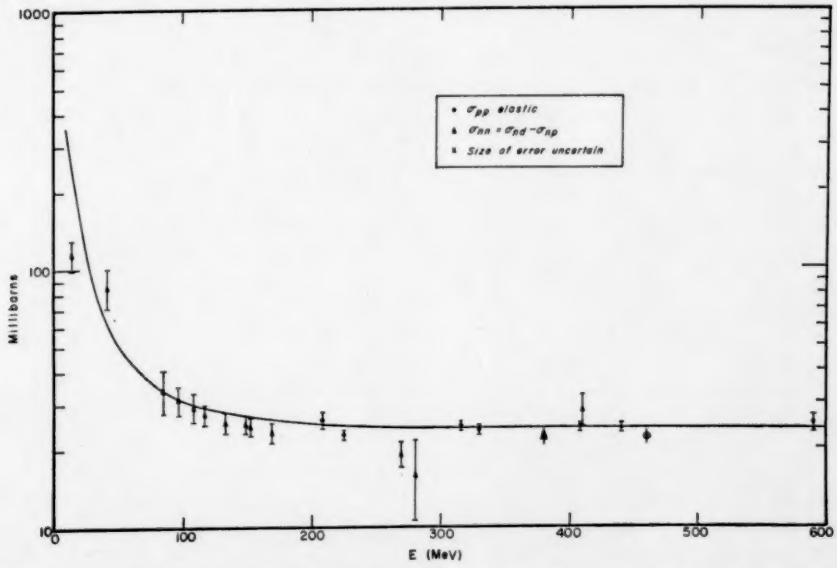

FIG. 11. Cross sections for (n, n) reactions (from HESS[4]). To be compared with (p, n) cross sections in Fig. 12.

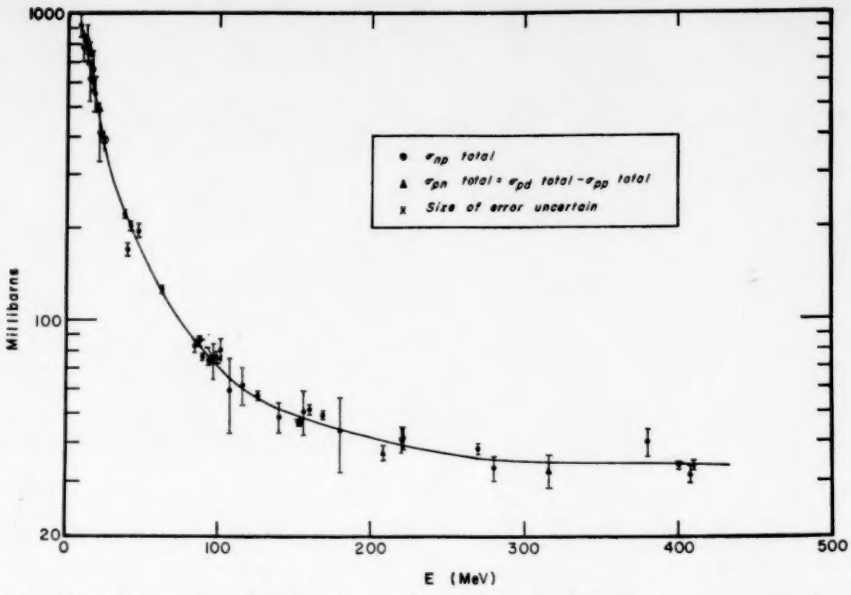

FIG. 12. Cross sections for (p, n) reactions (from HESS[4]). To be compared with (n, n) cross sections in Fig. 11.

energies, is applied as a correction factor to the better-known (p, pn) cross sections at corresponding energies. Thus, at 300 MeV the $(n, 2n)$ cross section has been estimated to be 24 mbarns.

Also theoretically, on the flat portion of the excitation curve, the yield is less for neutrons than for protons if exchange collisions occur. This is because a (p, n) exchange can lead to C^{11} by proton evaporation, but an (n, p) exchange cannot lead to C^{11}.[6]

No attempt has been made in the calculations to correct for this process. The calculated values for the $(n, 2n)$ cross sections should be viewed as approximations, and are more applicable above 100 MeV. However, the cross section has been measured for 90-MeV neutrons;[6] the results of three consecutive measurements are 21, 24 and 22 ± 4.0 mbarns. In a more recent report on a similar system using liquid scintillator, a cross section of 21–22 mbarns was used for neutron energies greater than 40 MeV; however, no reference for these values was cited.[7]

Near the $(n, 2n)$ reaction threshold, the cross section should be proportional to the square root of the energy excess.[8] The values of about 4.5 mbarns and 10 mbarns at approximately 23.5 and 27.5 MeV respectively[8] are consistent with theory.

Whether or not there is a peak in the $(n, 2n)$ reaction cross section in the region of 40 MeV depends on the importance of "intermediate-nucleus" reactions in relation to noncapture processes.[6] No such evaluations are included in this report; however, if such a peak exists, it will lead to an overestimation of the neutron flux density in this region.

In the region 22 ± 4 MeV there is a sharp (γ, n) resonance leading to the production of C^{11}.[10] The (γ, n) reaction cross section is of the order of 8 mbarns. At higher energies this cross section is less by two orders of magnitude. More than 60 per cent of these high-energy gammas will be absorbed in 1 cm of lead, so that it is easy to check on this reaction as a possible source of error.

VI. CONCLUDING REMARKS

Figs. 13 and 14 illustrate the difference in energy resolution between a 3×3-in. NaI(Tl) crystal and a plastic scintillator. The curves show the differential pulse-height distributions

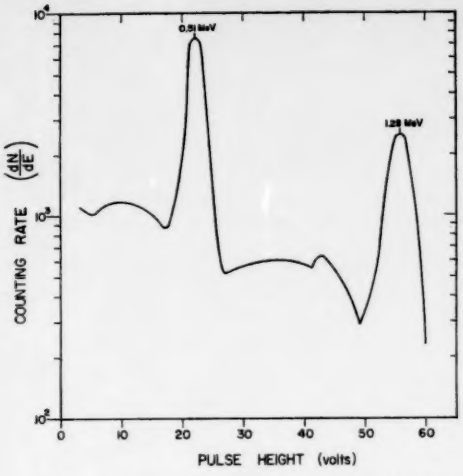

FIG. 13. Energy resolution with 3 × 3-in. NaI(Tl) crystal; the resolution (full width at half maximum) is 15% and 8% respectively at the two peaks.

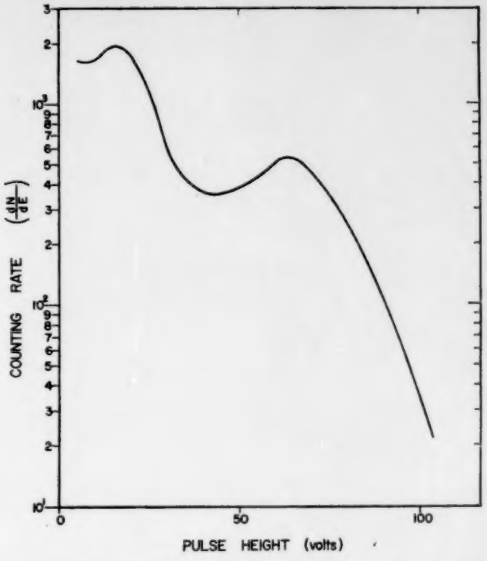

FIG. 14. Energy resolution for plastic scintillator with a Na^{22} source. Compare with Fig. 13.

from a Na^{22} source. The energy resolution of the system (full width at half maximum) with the NaI is 15 per cent and 8 per cent respectively on the two peaks. While such a system for C^{11} counting is being set up, a multichannel analyzer is of great assistance, but once it is set up a single discriminator is adequate. Gain is periodically checked by placing a gamma source in some standard geometry and making the necessary adjustments to produce a standard counting rate above the discriminator setting.

To summarize, a sensitive method of detecting high-energy neutrons and protons in the range 20 MeV to more than 400 MeV has been constructed with a counting error less than 15 per cent for flux densities of 1 neutron cm^{-2} sec^{-1}. Further experimental verification of the $(n, 2n)$ cross section would be desirable; however, sufficient information is available for most survey purposes.

Acknowledgments—The work presented in this report involved the help, advice, and co-operation of the entire Health Physics Group.

In particular the author wishes to thank Professor BURTON J. MOYER, Dr. ROGER W. WALLACE, H. WADE PATTERSON, LLOYD D. STEPHENS and T. M. JENKINS. The assistance of Dr. WILMOT N. HESS was invaluable in the preparation of the section on reaction cross sections.

A large share of the work, planning and evaluation was performed with the direct assistance of ALAN R. SMITH, for whose contributions the author is indeed grateful.

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NOTE

The Location of Lost γ-Ray or Neutron Sources

(*Received* 8 *December* 1959)

Introduction

CONTEMPLATION of the discussion of inverse square data by SLATER and MELVILLE[1], reveals a systematic method for precise location of lost sources emitting penetrating radiations, γ-rays or neutrons.

Method

The basic equation given by SLATER and MELVILLE is

$$\frac{1}{Vd^2} = \frac{1}{E}\left(1 + \frac{2a}{d} + \frac{a^2 + b^2}{d^2}\right) \qquad (1)$$

where V is the observed intensity, d is the presumed distance between effective centers of source and detector, E is a constant proportional to the source emission, a is the zero error in the distance measurement, and b the alignment error of the axis (see Fig. 1).

The previous authors noted that, in calibration of sources or detectors, both a/d and b/d tend to be small and hence dropped the last term of equation (1).

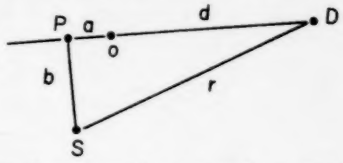

FIG. 1. Geometry for expression of inverse square law with detector at D, source at S and presumed location of source at P.

However, for the location of sources, we drop the terms in a and rewrite equation (1) as:

$$\frac{1}{Vd^2} = \frac{1}{E}\left(1 + \frac{b^2}{d^2}\right) \qquad (2)$$

and, from a plot or least squares analysis of $1/Vd^2$ vs. $1/d^2$, obtain b.

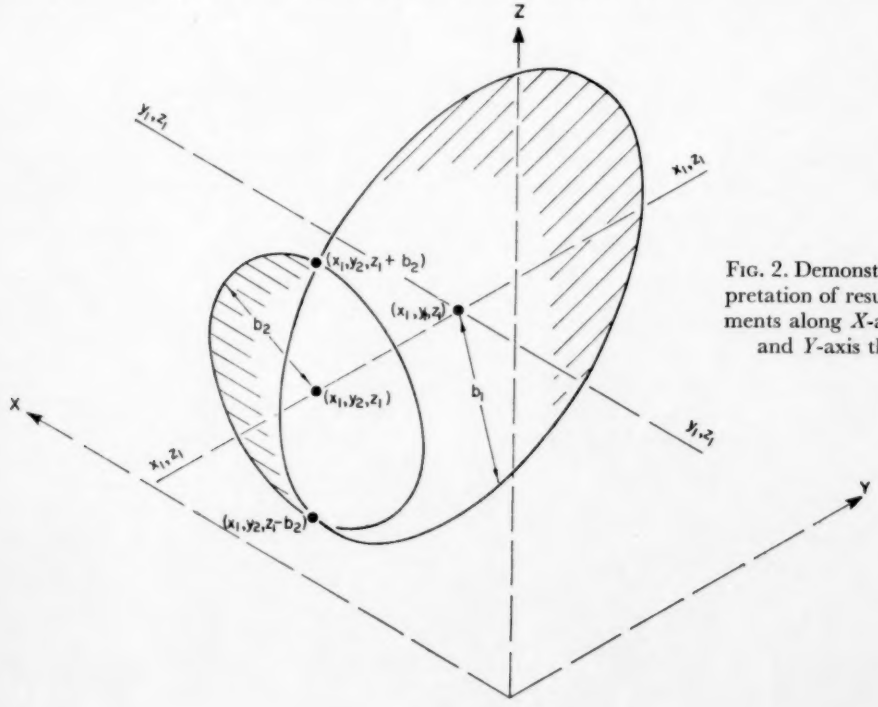

FIG. 2. Demonstration of the interpretation of results from measurements along X-axis through y_1, z_1 and Y-axis through x_1, z_1.

Successive sets of measurements along mutually perpendicular axes constitute the experimental data. If a source is within the test volume, the first set of measurements along an arbitrary X-axis defined by y_1 and z_1, should yield a maximum intensity at some point x_1 and calculations from this origin by equation (2) yield an alignment error estimate b_1 (see Fig. 2). Measurements along the Y-axis passing through x_1 and z_1, should yield a maximum at y_2 and a value b_2. Greater precision may be attained by further similar measurements along Z-, X- and Y-axes.

Discussion

Interpretations of the data are: from the first set, the source lies at a distance b_1 from the point (x_1, y_1, z_1) and near the YZ-plane defined by x_1; from the second set, the source lies at a distance b_2 from (x_1, y_2, z_1), near the XZ-plane, and likely near the points $(x_1, y_2, z_1 \pm b_2)$. Since successively smaller b-values are expected, a few sets of measurements should suffice.

The method should also be applicable for multiple lost sources because, when three axes have been traversed, more than one maximum reading should appear along at least one axis. Failure to observe more than one maximum renders some assurance of but one lost source. Inherent difficulties of this method include: the complexity added by scattering and shielding effects of objects in the test volume, and anisotropic properties or finite size of source or detector. Absorption and scattering is generally negligible for γ-ray and neutron sources in air but not in liquids. Location of intense α- or β-particle sources by bremsstrahlung detection may be feasible.

Summary

A systematic method of measurement and analysis for the location of lost sources of penetrating radiations has been outlined.

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Reference

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* Operated by Union Carbide Corporation for U.S. Atomic Energy Commission.

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NEWS

News Editor: J. J. FITZGERALD

Health Physics in the News

THE following items concerning Health Physics have appeared in newspapers, public press or technical journals during the last quarter.

Member of Health Physics Society made Chairman of ACRS

Professor LESLIE SILVERMAN of Harvard University, a charter member of the Society was chosen Chairman of the Advisory Committee on Reactor Safeguards. The Advisory Committee has the responsibility and authority of reviewing research (testing facilities as recently defined) and power reactors which may accumulate significant quantities of fission products. The Committee, after appropriate hearings on proposals to construct and to operate these reactors, makes their recommendations with respect to the proposals to the Chairman of the AEC.

AEC Health Physics Fellowship Selectors announced

Fellowships in Health Physics were granted to eighty-five applicants. The applicants had their choice of attending (upon acceptance by the University and the Fellowship Board) the Universities of Vanderbilt, Rochester, Washington, Kansas, California, Michigan and Harvard.

Fellowships were granted to five applicants under the AEC Special Fellowships for Advanced Training.

Employment column

Beginning with Volume 4, *Health Physics* will carry an employment column operated on a *confidential* basis through the Chairman of the Placement Committee. *Positions wanted* will be printed without identification of the subscriber except for a box number assigned by the Placement Committee. Replies to blind ads will be mailed to the Chairman of the Placement Committee in care of the assigned box number and then will be forwarded to the subscriber. *Positions open* may be printed with or without addresses at the subscribers request. The charges for ads on a single issue basis are as follows:

Position wanted—$0.25 per word ($4.00 minimum)

Position open—$1.00 per word ($15.00 minimum)

Ads mailed prepaid to Mr. J. A. LENHARD, Chairman, Placement Committee, Health Physics Society, Research and Development Division, U.S. Atomic Energy Commission, Oak Ridge, Tennessee, will appear in the next issue following receipt unless otherwise requested.

Radiological Health Curriculum Symposium planned

In view of the need for health physicists, the Public Health Service held a meeting in Washington to discuss the methods whereby an adequate number of "radiological safety physicists" may be educated. Professors in radiological sciences from several universities were invited to the meeting. Dr. FRANCIS WEBER, Chief of the Division of Radiological Health, made reference to the report of the Training Committee of the National Advisory Committee on Radiation which estimates the national needs exclusive of the AEC to be 1200 "radiological safety physicists" and 4000 "radiological technicians" by 1971. As a result of this meeting, a Symposium on Radiological Health Curricula is planned for late July or early August, 1960. At this symposium, papers will be presented, setting forth the basic elements that are believed necessary in the educational program for the development of future health physicists or "radiation safety physicists". Within two months following the symposium, it is hoped that the suggestions set forth at the meeting will be integrated into a report which will contain the recommendations wherein adequate action may be taken to accomplish this national need.

AEC proposes regulations for shipment of spent fuel elements

The Commission has invited comment on a proposed regulation to guard against accidental criticality, radiation exposure to individuals and the release of fission products during the shipment of irradiated fuel elements.

The AEC proposed regulation, Title 10, Code of Federal Regulations, Part 72—"Regulations to Protect Against Accidental Criticality and Radiation Exposure in the Shipment of Irradiated Fuel Elements"—was published in the Federal Register on March 17, 1960.

The fact that a number of reactors soon will be ready for refueling, and that construction of both domestic and foreign reactors has increased steadily, has created a need for irradiated fuel-element cask-design criteria and shipping procedures which will be acceptable from the radiological and criticality standpoints.

Under the proposed regulation each licensee making application to transport any number of irradiated fuel elements outside of his authorized location of use, or to deliver such material to a carrier, must submit to the Commission engineering drawings, experimental data and technical references showing that the cask to be used is designed for safety from the standpoint of criticality, heat transfer, shielding and structural integrity.

AEC formulates policy on land disposal of radioactive waste

The AEC has determined that regional disposal sites for permanent disposal of low-level packaged radioactive-waste materials shall be established as needed on State or Federal Government-owned land.

The publicly owned disposal installations would be operated by private contractors or licensees under strict Government controls or by the Federal or State Government and would be available to all users of radioactive materials.

Land requirements for disposal sites would not be large. On the basis of experience it is estimated that all such wastes generated between now and 1980 in the sixteen states of the Northeast area, could be safely disposed of in a 200–300 acre site.

The types of low-level wastes to which the Commission's policy applies include broken glassware, paper wipes, rags, ashes, animal carcasses, laboratory paraphernalia, etc. Low-level liquid wastes are treated and disposed of at their points of origin under existing controls and regulations.

Minnesota—AEC dispute

A dispute has arisen between the state of Minnesota and the AEC over state approval of reactor installations in addition to approval by the federal AEC.

In December, 1958, the state of Minnesota adopted a state board of health regulation which provides that no nuclear reactor may be operated in the state without the expressed approval of the Minnesota Board of Health. The regulation also stipulates that reactor construction cannot begin unless the Board has received a general description of the facility together with a hazard summary report, and has had an opportunity to object and to require changes in construction or operation deemed necessary in the interests of Public Health.

The right of a state to take such action under the federal constitution, in the absence of federal legislation specifically permitting such action, and in light of the Atomic Energy Act of 1954 conferring licensing authority on the AEC, has never been tested in court. However, a federal bill passed by Congress in September, 1959, provides for a gradual turnover of AEC regulatory authority in non-reactor areas to the states. This bill seems to contain the tacit assumption that the states do not automatically have such authority.

Nuclear standardization promoted by ASA activities

The proposed American Standards Association standard on a radiation symbol will be submitted at the International Standards Organization to be held in Geneva in May, 1960. A standard on radiation protection practices for uranium mines and mills has been developed. The proposed standard is being reviewed by the sponsor organization of the ASA committee.

The ASA N-6 Reactor Safety Committee reported their progress in developing operational standards at a meeting in New York City on January 12, 1960. Among the reports made at the meeting, progress in the following areas was noted:

(a) The third draft of a proposed standard on Site Selection Criteria has been completed.

(b) A proposed standard on Containment has been sent to sectional committee members for their review and comments.

(c) The Subcommittee on Failure Probabilities and Maximum Credible Accidents announced their decision to establish the "maximum design accident" as a basic criteria for their work. The "maximum design accident" is defined as the accident upon which containment design is based.

Ocean-bottom photographic analyses of Pacific disposal sites planned

A study of the underwater conditions at two waste-disposal sites in the Pacific Ocean will be made by the Cleveland Pneumatic Industries, Inc. of Cleveland, Ohio, under AEC contract.

At the site, approximately 52 miles west of San Francisco, disposal activities have been conducted since 1946. Approximately 21,000 concrete encased steel drums and 300 concrete boxes containing an estimate of 14,000 c (at the time of disposal) have been deposited at the site. At the second site, which is approximately 53 miles south of Santa Barbara in the Santa Cruz Basin, disposal activities since 1953 have involved an estimate of 2950 drums containing a total of 60 c.

Samples of marine life will be taken from the ocean floor by dredging the water above the disposal sites. Water-current measurements will be made including determinations of surface currents, dispersion and bottom-current velocity and direction. Radiochemical analyses will be made on specimens of seaweed sediment and marine life. Photographs will be taken of the ocean floors at both sites to ascertain the condition of the drums containing the disposed

radioactive materials. The results of these studies will be made available to the public.

Team of health physicists from ORNL study Yugoslav accident for IAEA

A dosimetry team from the Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, conducted a series of measurements at the Boris Kidrič Institute, near Belgrade, to determine the radiation doses of persons exposed at the time of the nuclear accident in October, 1958. The investigation was sponsored by the IAEA in agreement with the Yugoslav Government. French Nuclear Engineers from Saclay reconditioned the reactor, which had been left unaltered since the accident, and operated it during the experiments.

Six persons received exposures in the near-lethal or lethal range and were treated by the method of bone-marrow grafting at the Curie Hospital in Paris and all but one survived. This was the first successful application of bone-marrow transplanting in humans. The other two had received smaller doses and were treated in a hospital in Belgrade and also have recovered. Investigations as to the cause and consequences of the accident were immediately undertaken by the Yugoslav authorities and a report was published in the journal of the Boris Kidrič Institute, March, 1959.

Instrumentation in this study included threshold detector systems, proportional-counter neutron dosimeters, graphite–CO_2 ion chambers, metaphosphate glass detectors, other special counting devices and life-size human "phantoms" containing sodium solutions.

Scientists from other European Installations visited the site to observe the experiments. The results of the experiments will be published by IAEA.

A book called "Health Physics in Nuclear Installations," which consists of the proceedings of the 1959 Risø Symposium has been published by the O.E.E.C. Orders and enquiries from countries where sales agents have not yet been appointed may be sent to: O.E.E.C., Distribution and Sales Service, 33 rue de Franqueville, Paris 16e.

The Special Radiation Subcommittee of the Joint Committee on Atomic Energy is planning to hold public hearings on the subject "Radiation Protection Criteria and Standards: Their Basis and Use" to start in late May, 1960.

Summer School in Health Physics, Imperial College

A summer school in Health Physics (Radiation Protection) arranged by HUGH D. EVANS, B.Sc., Ph.D., A.Inst.P. (Lecturer in Radiation Hazards and Health Physicist), will be held in the Nuclear Technology Laboratories (Reader in Nuclear Technology: G. R. HALL, B.Sc., F.R.I.C.), forming part of the Department of Chemical Engineering and Chemical Technology, Imperial College.

The course will be of two weeks' duration starting on Monday, 4th July, 1960, and should be of interest to university radiation protection officers, industrial safety officers concerned with radiation and radioactive hazards, medical officers of health, factory inspectors and others concerned with radiation protection.

A university degree in science or medicine will normally be regarded as the minimum entrance qualification, but other suitably qualified persons will be considered.

The syllabus will include lectures on the following subjects:

Introductory nuclear physics
Units of radiation and radioactivity
Biological effects of radiation
Maximum permissible levels
Health Physics instrumentation
Biological monitoring procedures
Hazard control and laboratory design
Decontamination and waste disposal
Organization of radiation protection services
Regulations, national and international: Codes of Practice.

If there is a sufficient demand, it may be possible to provide a series of lectures on elementary nuclear physics, ionization, etc., prior to the main course.

At least one-third of the main course time will consist of practical work, and visits to other establishments in the London area will be arranged.

Residential facilities will be available in College hostels.

The fee for the course is 25 guineas exclusive of residence.

Applications for admission should be made to the Registrar, Imperial College, South Kensington, London S.W.7, from whom further information may be obtained.

The University of California will offer for the fifth year a number of courses and programs in the nuclear field during 1960. These include:

Nuclear Technology Survey—an executive review, July 18–22, 1960.

Criticality and Criticality Problems, June 20–July 1, 1960.

Basic Radiological Principles and Techniques, July 6–17, 1960.

Advanced Radiological Principles and Techniques, June 20–July 15, 1960.

The six-week program is divided into two- and four-week periods so that persons requiring only a basic knowledge of radiological principles may take only the first part. Those who require a more exhaustive knowledge may take the full six weeks, and persons already having completed a basic course elsewhere may start at the beginning of the four-week period.

In addition to industrial radioisotope users, personnel from state and local governmental groups and public health inspection and control groups will benefit from the two-week course. This provides a grounding in nuclear-science concepts necessary for intelligent administration and direction of activities involving radioisotopes and radiation.

For further information or application for enrollment, write to: Engineering and Sciences Extension, University of California, 2451 Bancroft Way, Berkeley 4, California.

A Symposium on Radioactivity in Man, Measurement and Effects of Internal Gamma Ray Emitting Radioisotopes, was held at Vanderbilt University, on April 18 and 19, 1960. Program Chairman was GEORGE R. MENEELY, School of Medicine, Vanderbilt University, Nashville 5, Tennessee.

The Fifth Annual Meeting of the Health Physics Society will be held at the Hotel Statler Hilton in Boston, Massachusetts, Wednesday through Friday, June 29, 30 and July 1, 1960. The preliminary program is:

Wednesday, June 29

Afternoon: Field trips

Evening: Symposium on Sea Disposal (invited papers)

Chairman, E. G. STRUXNESS, Oak Ridge National Laboratory, Oak Ridge, Tennessee

Contributed Papers on Dosimetry

Chairman, W. T. HAM, Medical College of Virginia, Richmond, Virginia.

Thursday, June 30

Morning: Symposium on Effect of NCRP Recommendations on National Life (invited papers—Labor, Legal, Medical and Insurance)

Chairman, H. P. YOCKEY, Aerojet-General Nucleonics, San Ramon, California

Afternoon: Contributed Papers on Air Cleaning, Atmospheric Contamination and Fallout

Chairman, L. SILVERMAN, Harvard University, Cambridge, Mass.

Contributed Papers on Health Physics Instruments

Chairman, J. B. H. KUPER, Brookhaven National Laboratory, Upton, L.I., New York

Evening: Banquet speaker, Secretary of Health, Education and Welfare—ARTHUR S. FLEMMING, "The Federal Radiation Council"

Friday, July 1

Morning: Symposium on Recent Case Histories in Applied Health Physics Programs (invited papers)

Contributed Papers on Internal Radiation Hazards and Internal Dosimetry

Chairman, L. S. SKAGGS, Argonne Cancer Research Hospital, Chicago, Illinois

Afternoon: Contributed papers on Environmental Hazards of Radiation and Reactor Hazards (also invited paper)

Chairman, H. F. SCHULTE, Los Alamos Scientific Laboratory, Los Alamos, New Mexico

Contributed Papers on General Topics in Health Physics

Chairman, A. L. CARSTEN, Brookhaven National Laboratory, Upton, L.I., New York

Registration will be held at the Statler Hilton from 10.00 a.m. until 5.00 p.m. on Wednesday, June 29. Registration will reopen on Thursday, June 30, at 9.00 a.m.

American Board of Health Physics for certification in Health Physics

At its organizational meeting on March 11, 1960 the American Board of Health Physics set up the procedures by which it will provide Health Physics Certification of radiation-protection experts throughout the United States. Under the procedures each candidate for certification will have to pass a written examination, but this examination may be waived for especially well-qualified persons who apply before December 31, 1960.

The American Board of Health Physics was established last October by the Health Physics Society, but it will operate as an independent body. At present the Board is composed of five members, all nominated by the Health Physics Society. Other organizations will be asked to nominate additional members in the near future. The present members of the Board are ELDA E. ANDERSON, Oak Ridge, Tennessee; J. W. HEALY, Richland, Washington; J. S. LAUGHLIN, Scarsdale, New York; W. A. MCADAMS, Scotia, New York and L. S. TAYLOR, Bethesda, Maryland. Mr. MCADAMS is Chairman of the Board.

The examinations, both written and oral, will be conducted by an Examination Panel which is still to be selected. Membership in the Health Physics Society is not required for certification, nor for membership on the Board or on the Examination Panel.

It is expected that the first certificates of certification will be issued about July 1, 1960.

Application blanks for certification may be obtained from Dr. ELDA E. ANDERSON, Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee. An application fee of $15 must be paid when the application is submitted to the Board and will not be returned if the applicant fails to qualify. An additional fee of $10.00 will be charged when certification is approved.

Perhaps in the near future and as the need arises additional provisions can be made for Health Physics Certification in other countries.

Qualifications for a Certified Health Physicist

(1) *Academic.* The applicants must have a Bachelor's degree in either a physical or biological science with a major in physics or a major in chemistry, engineering or biology with a strong minor in physics. In exceptional cases, persons who have demonstrated adequate knowledge of Health Physics but who are deficient in the formal requirements above may, at the discretion of the Board, be permitted to substitute experience for academic requirements.

(2) *Experience.* All applicants must have had at least *five* years of *responsible* professional experience in Health Physics. Experience in training or as a technician does not count. One year of graduate work in a field closely related to Health Physics may be substituted for a year of experience up to a maximum of three years. Considerable weight will be given to recommendations from reliable persons relative to calibre of applicant and responsible nature of his work.

(3) *Professional.* All applicants must be engaged in the professional practice of Health Physics a substantial portion of their time.

(4) *Written Report.* All applicants must be capable of making a satisfactory Health Physics evaluation on several installations or operations of which those listed below are examples. The Board after examination of the application for certification will normally request reports on at least two such evaluations made personally by or under the supervision of the applicant. In the case of applicants engaged primarily in research and development in the field of Health Physics, reports of these activities which have appeared in open literature may be submitted alternatively.

(a) Radiographic installation—industrial or medical
(b) Fluoroscopic installation
(c) Therapy installation
(d) Radioisotope laboratory
(e) Air and water sampling and environmental survey
(f) A radioisotope chemical separation plant
(g) Reactors
(h) Major decontamination operation
(i) Accelerators
(j) Others

(5) *Oral Examination.* After Dec. 31, 1960, written and/or oral examinations by at least three members of the Examination Panel will be mandatory for applicants who have satisfied all other requirements. Persons certified without examination prior to Dec. 31, 1960, must be approved by unanimous vote of the American Board of Health Physics.

(6) The Board of Directors of the Health Physics Society reserves the right to change the above requirements as experience dictates, no such changes to be retroactive and all such changes to be publicized well in advance of their inauguration.

A radiation exposure has been reported by AEC at Oak Ridge, Tennessee

On March 8, an ORNL employee received a radiation exposure to the skin above levels established for routine occupational activities. The o erexposure occurred during a scheduled clean-up of a cell used for handling radioactive materials. The exposure was from cerium-144 and praseodymium-144. No significant inhalation occurred.

Based on evaluation of the radiation surveys and film-badge data, the exposure is now estimated as 80 rads to body skin and 100 rads to face skin. Permissible occupational exposure for the skin, as given in the NCRP, is 6 rads per 13 weeks. The exposure to the lens of the employee's eyes is estimated at 5 rads as compared to limits of 3 rads in 13 weeks. The employee's exposure to penetrating radiation (gamma) is estimated to be 2.5 rads, compared to the maximum permissible occupational exposure of 3 rads in 13 weeks.

Health Physics news becomes appropriately international

Since one of the objectives of the Society is the promulgation of effective and adequate measures of radiation protection and control through the medium of the Journal on an international basis, it seems appropriate at this time that we extend our news coverage to provide active and effective international representation. Such broadening of coverage should be beneficial to all the members of the Society. To achieve the above objective, representatives from 18 nations were selected. There have been no refusals to participate in this program. Members from eleven of these countries have replied in the affirmative.

The following persons have, at this time, indicated their willingness to participate in the program.

R. R. Boulenger, Belgium
John Neil, Canada
B. C. Christensen, Denmark
H. J. Dunster, England
Francis Duhamel, France
Laurence Collery, Ireland
Enrico Via, Italy
Toshio Aoki, Japan
Leiv Berteig, Norway
B. G. Lindell, Sweden
Perihan Cambel, Turkey

Health Physics activities in Italy—Enrico Via

The Italian Health Physics Association was established in 1958. At the present time the Association has fifty-eight members, consisting of physicians, engineers, chemists, biologists, radiologists, etc. and representatives of industrial groups who are connected with atomic-energy operation. The physicians associated with Italian universities represent a relatively large portion of the membership. The journal *Minerva Nucleare* is the official organ of the Association.

The Italian Health Physics Association is well integrated with similar European associations, especially the associations of the bordering nations of France and Switzerland. During 1959 a meeting was held with a group of French specialists in Grenoble, France. The last annual meeting was held in Bologna at the Physics Institute (the Italian "Center of Dosimetry").

Health Physics activities in Denmark—B. C. Christensen

In Denmark, an emergency plan for the atomic-energy installation is being prepared. This plan will be summarized in this column in a future edition of the Journal.

The activities of health physicists in Scandinavia are co-ordinated such that they meet every six months to discuss problems of mutual concern. Two conferences were held in 1959. The first conference at Oslo, Norway, was held in June and the second at Stockholm, Sweden, in November. During the conference in June, 1959, the experts discussed the norms which might be applied to an evaluation of the health hazard due to radioactive fallout. They agreed that ICRP's recommendation should form the basis of such evaluations.

The Scandinavian experts hold the view that, in the light of our present knowledge, there is no reason for the Scandinavian Health Authorities to make special provisions as long as the levels recommended by ICRP are not exceeded. If the recommended values for individuals, population groups or the population as a whole are exceeded over a relatively long period of time, the Health Authorities should consider appropriate measures, taking into account various technical, dietary and economical factors.

At their first conference, the Scandinavian experts attempted to arrive at a preliminary estimate of the somatic hazard to the population from radioactive fallout. It was agreed that the maximum permissible concentration of Sr^{90} in the diet of the population should correspond to an average of 75 S.U.

On the basis of available radioactivity measurements and data on the dietary habits in the Scandinavian countries the experts estimate the average Sr^{90} intake with the food in Scandinavia in 1959 to be between 5 and 25 S.U. (A Sunshine Unit is equal to 1 $\mu\mu$c of Sr^{90} per g of Ca.)

Although it is difficult to convert these figures into a skeleton and a bone-marrow dose, values fall considerably below the accepted tolerance dose—similarly if the dose from external radiation is added. The experts estimate that the average genetic dose to the population is about 5–10 mrems, varying between 3 and 20 mrems.

The Scandinavian experts arrived at the conclusion that radioactive fallout in Scandinavia, as recorded up to the present, does not require special precautions to be taken by the Health Authorities. An evaluation of the relation between fallout activity and contamination of food products requires detailed agricultural and veterinary-medical investigations which must be carried out in every country so that special local factors can be taken into account.

The experts thus consider it urgent that Health Authorities in their respective countries support investigations which will clarify these problems, not only in view of the fallout situation of to-day, but also with regard to the possibility that testing of nuclear weapons might be resumed and, last but not least, in view of the increase in the level of contamination as a consequence of the peaceful uses of atomic energy.

Health Physics experts from Denmark, Finland, Norway and Sweden gathered for a second conference in Stockholm on November 23–26, 1959, to discuss questions of radiation hygiene. The discussions centered around problems of the radiation exposure of the Scandinavian people due to radioactive fallout—moreover, questions arising in connection with potential reactor accidents were discussed.

As a consequence of nuclear-weapon tests in the autumn of 1958, considerable amounts of fission products were injected into the stratosphere. The concentration of fission products in the air at ground level and in precipitation increased shortly after the

tests, and continued to increase until May 1959. During the following months, the concentration of radioactive isotopes in the air at ground level and in precipitation decreased all over Scandinavia, and by the end of October, 1959, the values measured at various locations were only about 1/10 to 1/100 of those recorded in May, 1959.

The radioactivity measurements carried out at different altitudes in Norway and Sweden support the view that the previously assumed pattern of the global fallout (a mean residence time of the radioactive dust in the stratosphere of about 5 years) is wrong, at least as regards fallout from nuclear-weapon tests in northern latitudes. According to the above-mentioned measurements, the mean residence time over Scandinavia of debris injected at northern latitudes seems to be of the order of 4–6 months. This discrepancy causes considerable uncertainty in the evaluation of the mechanism determining the transport of debris from the stratosphere to the surface of the earth.

The quantity of radioactive material now deposited on the ground over Scandinavia is on the average twice that observed in the autumn of 1958.

The increase in the amount of fission products accumulated on the ground in Scandinavia has led to an increase in the radioactive contamination of water and foodstuffs. In different parts of Scandinavia the means for 1959 observed in some foodstuffs vary as follows:

For Sr^{90} in milk—5–25 S.U.

For Cs^{137} in milk—20–100 $\mu\mu c/l$.

For the total activity in drinking water—5–20 $\mu\mu c/l$.

United Kingdom developments in health and safety legislation—H. J. DUNSTER

The last few months have seen several major developments in the law relating to health and safety aspects of nuclear energy and radioactive materials. The Nuclear Installations (Licensing and Insurance) Act received the Royal Assent last year and is expected to come into effect in the Spring of 1960. It provides that nuclear installations, a term including reactors and plants for preparing and processing their fuels, may operate only in accordance with licences granted by the Minister of Power, or the Secretary of State for Scotland. The Act also lays down financial arrangements for dealing with claims, and limits such claims to those made within a period of 30 years from the date of the incident or operation giving rise to the claim. A duty is put on the licensee to secure that no ionizing radiation causes any hurt to any person or property on or off the licensed site.

Two further developments occurred at about the end of 1959. A Bill was introduced to modernize and improve the control over the discharge of radioactive waste, and draft regulations were published on the use of sealed radioactive sources and X-ray sets in factories. The main provisions of the Bill, the Radioactive Substances Bill, 1959, are to require the registration of all premises where radioactive materials are used or stored and to prohibit the disposal or storage of radioactive waste except in accordance with written authorizations granted by the Minister of Housing and Local Government. At the same time the radioactivity of the waste is removed from the scope of the conventional public health legislation, such as the Rivers (Prevention of Pollution) Act, 1951. This section of the Bill will thus take the control of radioactive wastes out of the hands of the local authorities and make it a responsibility of central government. A White Paper accompanying the Bill and called "The Control of Radioactive Wastes" makes it clear, however, that local disposal is still to be encouraged and that the proposed national disposal service will be available only when conventional means of disposal cannot be used with safety.

The draft regulations, the Factories (Ionizing Radiations) Special Regulations, lay down rules for operating procedures, monitoring, medical supervision and maximum permissible doses to be applied wherever sealed radioactive sources or machines generating radiations such as X-rays are used in factories. These rules emphasize the importance of putting primary reliance on shielding rather than on the use of distance or the control of operating time. The Regulations will be administered by the Ministry of Labour.

All the documents referred to can be obtained from H. M. Stationery Office, York House, Kingsway, London, W.C.2.

Health Physics activities in Norway—LEIV BERTEIG

Recently, a committee consisting of persons from organizations and associations connected with atomic energy, shipping, trade, health agencies, etc. discussed and initiated investigations on problems concerned with nuclear ship safety, such as:

(1) Waste disposal
(2) Radiolytic gas production and "off gassing."
(3) Maneuvering characteristics of large conventional turbine tanker to establish design criteria for the nuclear tanker
(4) Construction of the nuclear ship
(5) How to shelter the reactor in case of collision
(6) Emergency equipment
(7) Particular efforts to prevent fire

(8) Removal of the reactor if the ship goes down
(9) Harbor and navigational problems.

The unofficial committee is now relieved by a new governmental committee consisting of only seven persons with deputies. Represented in the committee are:

Board of Trade
The State Institute of Radiation Hygiene
Harbor Directorate
Institute for Atomic Energy
Ship officers and crew organizations
Det norske Veritas (a ship control organization)
Ship owners association

The committee has been given the following mandate:

(a) To set forth the Norwegian position at the international conference May/June, 1960, in London, about safety for man on sea, considering nuclear ship construction, fitting up, equipment, crew, operation, etc. The committee will also analyze the view points and positions of the other member countries at the conference.

(b) To develop a policy and if possible give temporary directions for nuclear ship construction, equipment, crew, operation, together with the measures and security efforts that are considered necessary to ensure satisfactory conditions for crew, passengers and persons who during their work enter the ship, such as the loading and unloading workers, etc.

(c) To determine whether nuclear ships may be allowed to traffic Norwegian waters and call at Norwegian harbors and, in case of such allowance, give statement as to which waters and harbors this allowance is given.

Reactor accidents—A committee has discussed the state of "standby" in Norway in case of reactor accidents. Responsibility and fields of work will be determined in a new law concerning ionizing radiation.

I^{131} *in milk*—The State Institute of Radiation Hygiene has worked out a method for the rapid determination of I^{131} in milk. This method is based on the application of a counter with a one-channel discriminator. With this method, amounts down to 2 mμc I^{131}/l. milk can be traced without delaying the work in the dairy.

A LIST OF PAPERS TO BE PUBLISHED IN FUTURE ISSUES

J. H. SPAA: A rapid-indicating instrument for the stepwise measurement of airdust radioactivity

G. M. DUNNING: Cobalt-60 bombs

SOHEI KONDO: Neutron response of silver-activated phosphate glass

E. A. EDELSACK, W. E. KREGER, W. MALLET and N. E. SCOFIELD: Experimental investigation of thick target bremsstrahlung radiation produced by electrons of 1.00, 1.50 and 2.00 MeV

C. A. ZIEGLER and D. J. CHLECK: Latent-image fading in film-badge dosimeters

G. M. DUNNING: Shorter-lived fission products in fallout

C. SHARP COOK: The energy spectrum of gamma radiation from fallout

J. A. DAVIS, LOREN C. LOGIE, E. M. ROBINSON and B. S. HEATHINGTON: A correlation analysis of the relationship between weather variables and atmospheric radioactivity

D. A. CROSSLEY, JR. and M. E. PRYOR: The uptake and elimination of cesium-137 by a grass-hopper—*Romalea microptera*

ERRATUM

Vol. 2, No. 3, p. 312. The following correction has been received for the news item *Federal Radiation Council*. The second line onwards should read:

"... the President also directed that the Department of Health, Education and Welfare intensify its radiological health efforts and have primary responsibility within the executive branch for the collation, analysis and interpretation of data on environmental radiation levels such as natural background, radiography, medical and industrial use of isotopes and X-rays, and fall-out, so that the Secretary of Health, Education and Welfare may advise the President and the general public."

END OF VOL.